Concentrations and emission rates of indoor VOCs—a comparative study between Singapore and European office buildings

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ABSTRACT

This study highlights the presence of dominant source/s of several VOCs and their geographical similarities and differences within European Union (EU) and Singapore buildings. Concentrations and emission rates of 2-methylpentane and heptane are higher in EU buildings as compared to the ones in Singapore. Also, lower alkanes are observed to be higher in levels in EU buildings as opposed to the higher alkanes, which are more dominant in Singapore buildings with statistical differences approaching significance. Benzene and toluene concentrations and emission rates are significantly higher in ETS-free Singapore buildings. There are no differences in VOCs (1,1,1-trichloroethane and limonene) related to occupants suggesting similar use of consumer products. However, differences in emission rates of limonene approach significance which maybe related to wood products commonly used in European buildings. Higher emission rates for isoprene in EU buildings could be due to building materials choice instead of bioeffluents emissions.

INDEX TERMS

VOCs; Concentrations; Emission rates; European Union; Singapore; Office buildings

INTRODUCTION

The occurrence of SBS symptoms in Singaporean office workers is comparable to that of the European Union (Roulet *et al.*, 2002). Although agents responsible for these complaints remain largely unknown, it has been documented that volatile organic compounds (VOCs) may play a role in producing symptoms such as eye and airway irritation, headache, nausea and dizziness and may play a role in the high prevalence of SBS symptoms. Indoor environment quality studies were performed in six European countries within the framework of a European research programme (Bluyysen *et al.*, 1995) and also in Singapore (Sekhar *et al.*, 2001). While the comparison on ventilation, indoor environment quality and climate between the EU and Singapore buildings has been reported elsewhere (Roulet *et al.*, 2002), this paper focuses on the indoor VOCs.

All buildings investigated are office buildings. The buildings were described in detail in their respective reports (Bluyysen *et al.*, 1995; Sekhar *et al.*, 2001). These buildings are located in various climates. The Nordic climate includes Danish, Norwegian and Finnish buildings (18 buildings); Continental Europe includes 20 buildings from France, Germany and Switzerland; Oceanic Europe includes 12 buildings from the UK and Holland; Mediterranean Europe includes six Greek buildings while the tropical climate involves eight Singapore buildings. All European buildings were investigated in winter, during heating seasons. The climatic differences between the European climates are not large, except for Greece (Roulet *et al.*, 2002). The main differences between buildings investigated in Europe and Singapore are shown in Table 1.

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Table 1 Main differences between the buildings investigated in Europe and Singapore

Europe	Singapore
Heating mode	Cooling mode
Temperate or cold climate	Tropical climate
Recirculation rate from 0 to 90%	High recirculation rate (about 90%)
Large outdoor air flow rate per occupant or square metre floor area	Reduced outdoor air flow rate per occupant or square metre floor area
31 ± 5% smokers on average in buildings	No smoking allowed
Mean building age: 17.9 years	Mean building age: 3.2 years
Locality : 76% suburb or downtown	Locality: 87.5% suburb or downtown

METHODS

For the European study, VOCs were collected in conditioned Tenax TA adsorbent tubes by active sampling. For sampling, 10 tubes were provided for each building. The adsorbent tubes from all the countries were then transported to a central laboratory for analysis. The VOCs were thermally eluted by an automated thermal desorber and transferred to a gas chromatograph. VOCs were quantified using flame ionization detection and qualified using mass spectrometry. Quantification of individual VOCs was by using Toluene as a reference.

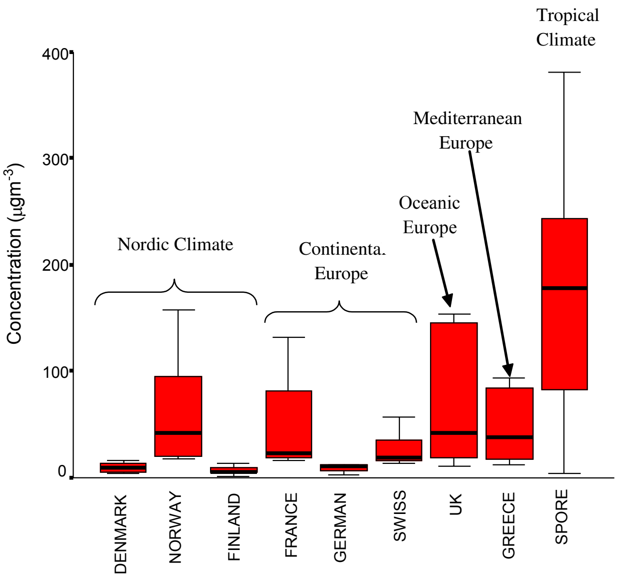
For the study conducted in Singapore, VOCs were collected on conditioned multisorbent tubes by active sampling. For each building, several floors which represent 20% of the building height were selected. Within each measurement floor, five indoor locations and an ambient location were selected for sampling. The analysis of collected VOCs is almost similar to the method except that quantification is by mass spectrometry. Also, individual VOCs were quantified using their own response factors. However, to facilitate comparison, the values in this study were converted to toluene-equivalent.

Fifteen VOCs which were the most abundant compounds were targeted for each building (Bluyssen *et al.*, 1995). Area specific emission rates (SER) of the target VOCs were computed using a simplified mass-balanced equation described elsewhere (Zuraimi *et al.*, 2003).

RESULTS

Samples collected within the EU and Singapore buildings revealed a total of 29 common VOCs. The number of frequent VOCs ranges from two to seven compounds between the EU countries. Among these, the common VOC that appears in all buildings across the EU countries is toluene. For Singapore buildings, however, 20 VOCs appear in every building. VOCs with mean concentrations higher than 100 $\mu g\ m^{-3}$ in the EU countries include *n*-hexane (Switzerland and Norway) and limonene (Norway). In Singapore buildings, two aromatic VOCs (toluene, *m*-/*p*-xylene) have mean concentrations higher than 100 $\mu g\ m^{-3}$.

Concentration of toluene is the highest in Singapore among all buildings studied (see Figure 1). Also, it is the most abundant and frequent VOC that is common among EU and Singapore buildings. Toluene is then used to evaluate whether there are significant differences in concentrations among the different EU climates. This difference is not significant ($p = 0.112$). Therefore, comparisons are made between EU countries and Singapore. Their *p*-values are tabulated in Table 2. From Table 2, it is observed that 2-methylpentane and *n*-heptane are significantly higher in EU than in Singapore buildings with the lower alkanes (3-methylpentane, hexane) approaching significance. The aromatic VOCs like benzene, toluene, *m*-/*p*-xylene and benzaldehyde are significantly higher in Singapore than in EU buildings. There are no significant differences for VOCs associated with building occupants like limonene, 1,1,1-trichloroethane, 2-propanone and isoprene.



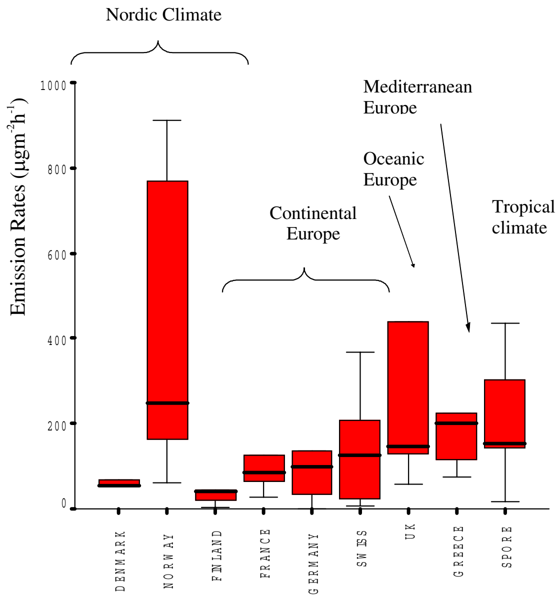


Figure 1 Concentrations and emission rates (SER) of toluene in EU and Singapore buildings

Table 2 Mean concentration (µg m^{-3}) of target VOCs for EU and Singapore buildings

Compound	EU countries								EU mean	S'pore mean	*p*
	Switzerland	Germany	Denmark	UK	Greece	France	Finland	Norway			
2-Methypentane	9.7	3.2	3.0	23.6	17.7	15.0	1.0	41.2	14.3	6.4	*0.01**
3-Methylpentane	24.7	2.6	10.0	33.2	15.4	–	–	51.5	22.9	5.7	0.09
n-Hexane	201.2	–	3.5	36.8	15.0	20.2	3.2	148.3	61.2	34.6	0.13
n-Heptane	33.8	2.6	4.0	20.6	10.1	10.7	2.0	77.5	20.2	14.7	*0.04**
n-Nonane	20.8	1.4	–	–	77.7	–	2.0	–	25.5	8.3	0.34
n-Decane	12.5	3.4	6.0	12.8	–	3.6	–	9.0	7.9	24.8	0.17
n-Undecane	5.5	4.0	3.0	35.5	–	13.3	–	32.0	15.5	32.9	0.22
Isoprene	6.4	1.6	7.7	4.0	13.4	–	–	–	6.6	10.5	0.33
Benzene	–	3.8	4.4	17.1	24.7	18.6	2.8	30.8	14.6	87.1	*<0.001**
Toluene	26.7	12.2	10.0	68.8	47.3	49.1	7.2	62.3	35.5	287.3	*<0.001**
o-Xylene	7.4	3.5	2.7	14.5	19.2	13.2	–	3.0	9.1	43.4	0.07
m-/*p*-Xylene	20.6	8.0	5.8	17.9	69.4	32.7	6.3	11.2	21.5	143.0	*<0.01**
Ethanol	12.9	8.8	5.0	5.2	–	45.5	–	–	15.5	17.2	0.16
1-Butanol	62.1	3.1	4.0	–	–	9.5	2.0	–	16.2	4.4	0.15
Methylcyclopentane	31.8	–	–	31.0	–	–	–	70.7	44.5	8.1	0.18
Methylcyclohexane	30.2	2.9	5.0	18.3	–	5.4	1.3	54.8	16.9	36.4	0.39
1,1,1-Trichlorethane	7.0	–	–	21.6	12.6	20.9	2.0	–	12.8	36.1	0.62
2-Propanone	25.0	19.6	6.2	22.2	26.5	20.0	1.5	5.7	15.8	16.5	0.85
Benzaldehyde	3.7	2.5	3.3	5.3	1.2	8.2	–	–	4.1	29.0	*<0.001**
Limonene	23.1	14.8	3.3	30.5	26.1	50.2	2.0	186.5	42.1	65.1	0.78

* $p < 0.05$.

The SER values are tabulated in Table 3. Emission rates of VOCs significantly higher in EU than in Singapore buildings include 2-methylpentane, *n*-heptane and isoprene. Benzene and toluene SER values are significantly higher in Singapore than in EU buildings.

Table 3 Mean SER ($\mu g\ m^{-2}\ h^{-1}$) values of target VOCs for EU and Singapore buildings

Compound	EU countries								EU mean	S'pore mean	*p*
	Switzerland	Germany	Denmark	UK	Greece	France	Finland	Norway			
2-Methypentane	73.9	26.0	15.7	88.1	127.0	43.3	7.4	216.8	74.8	13.6	*0.01**
3-Methylpentane	203.4	14.3	52.4	133.4	103.6	–	–	272.3	129.9	13.7	0.05
n-Hexane	1657.3	–	21.7	156.1	108.4	48.8	18.0	777.5	398.2	66.1	0.06
n-Heptane	231.3	5.6	16.0	95.8	34.5	33.6	11.6	387.0	101.9	30.9	*0.02**
n-Nonane	171.7	13.2	–	–	293.7	–	13.4	–	123.0	22.7	0.17
n-Decane	80.4	42.9	54.8	40.9	–	14.8	–	73.6	51.2	70.4	0.13
n-Undecane	28.5	50.0	16.9	125.5	–	14.7	–	261.8	82.9	69.9	0.12
Isoprene	25.0	19.7	46.4	46.7	101.9	–	–	–	47.9	22.9	*0.02**
Benzene	–	31.6	21.2	73.8	183.5	50.9	18.0	177.7	79.5	200.3	*0.02**
Toluene	181.5	119.4	56.4	330.1	375.1	127.6	42.4	400.4	204.1	393.0	*0.01**
o-Xylene	61.6	41.5	15.1	47.3	119.3	29.6	–	16.0	47.2	83.7	0.25
m-/*p*-xylene	150.8	90.1	29.7	67.3	376.4	92.8	38.6	73.7	114.9	207.4	0.16
Ethanol	18.6	25.7	26.2	39.0	–	161.0	–	–	54.1	41.9	0.09
1-Butanol	411.9	9.2	22.7	–	–	49.2	7.7	–	100.1	12.8	0.16
Methylcyclopentane	261.5	–	–	126.6	–	–	–	369.3	252.5	19.2	0.12
Methylcyclohexane	205.6	15.3	8.4	89.8	–	18.0	7.3	283.1	89.6	68.9	0.47
1,1,1-Trichlorethane	57.1	–	–	102.8	112.2	55.7	9.7	–	67.5	45.7	0.99
2-Propanone	166.3	182.3	40.3	106.4	219.3	59.0	9.0	39.3	102.7	54.2	0.13
Benzaldehyde	21.3	16.9	19.6	38.2	14.9	21.7	–	–	22.1	61.8	0.12
Limonene	120.4	157.9	19.5	139.0	98.5	191.1	18.1	659.3	175.5	122.1	0.06

* $p < 0.05$.

Table 4 Environmental parameters of EU and Singapore buildings

	S'pore mean (±)	EU mean (±)	Difference S'pore – EU	*p*
ACH	0.58 (±0.34)	2.43 (±1.89)	–1.85	<0.001
Temperature	22.8 (±1.3)	23.0 (±0.5)	–0.2	0.67
Number of persons per room	23.4 (±19.6)	15.0 (±16.9)	8.4	0.27

DISCUSSION

Generally, it is observed that in EU buildings, concentrations of lower alkanes are higher than in Singapore buildings. Probable sources for 2-methylpentane includes chipboard, gypsum board, insulation foam, floor coverings, wallpaper and even traffic combustion while possible of heptane are floor coverings and varnishes. However, these sources are not unique in both EU and Singapore buildings. Interestingly, 2-methylpentane concentration is higher in EU and Singapore buildings older than 2 years as compared to new ones (19.5 versus 2.5 $\mu g\ m^{-3}$ and 3.4 versus 2.4 $\mu g\ m^{-3}$, respectively). Unlike the lower alkanes though, undecane has concentration higher in Singapore buildings. This difference, however, is not significant.

Concentrations of ETS-related aromatic compounds are higher in Singapore. It is interesting because smoking within Singapore buildings is not permitted as opposed to those in EU. Therefore, aromatic compounds in Singapore buildings may originate from other sources which can be dominant. This is alarming considering these compounds can affect health in a significant way. Benzene, toluene and xylene isomers are either known or suspected carcinogens. The geographical difference in emission rates revealed higher levels in EU buildings for three VOCs (isoprene, 2-methylpentane, heptane) as opposed to two for differences in concentrations. This could be due to the significantly higher ACH (Table 4) in EU buildings as compared to that in Singapore. Despite this, benzene and toluene SER values are higher in Singapore buildings. Indeed, this highlights their indoor dominant source.

Benzene is used in adhesives, spot cleaners, alkyd paints, paint removers, particle board, furniture waxes, carpeting and electrical insulation. Toluene is used for rubbers, wallpaper, floor coverings, jointing compounds, caulking compounds, vinyl floor coverings, paints, chipboard, solvent- and water-based adhesives, inks and detergents. However, these sources are not unique in both EU and Singapore buildings. Possible reasons for the elevated levels in Singapore buildings could be due to the specific choice of materials, which in Singapore might contain more of these compounds and occupant activities in Singapore buildings.

Differences in behaviour or choice of consumer products between occupants in EU and Singapore buildings could not be established. Comparisons of concentrations of 1,1,1-trichloroethane (TCA) and limonene which have mainly indoor sources and are commonly found in many cleaning products, air fresheners and stationery items revealed no significant differences. However, differences in emission rates of limonene approach significance which maybe related to wood products that are commonly used in European buildings (Molhave *et al.*, 2000) or even ETS. The higher emission rates of heptane and 2-methylpentane are due to the widespread use of solvents (Bluyssen *et al.*, 1995), which typically contain more of these compounds.

Occupant related 2-propanone and isoprene (both human bioeffluents) levels between the two climates are rather conflicting. Firstly, there is no difference in concentration levels, which suggests that the dominant source is from the occupants. Now, there is no geographical difference in the occupant density. Since the ACH is higher in EU buildings, it is natural to assume that emission rates of the presumed dominant human source to be reduced to the same degree for both VOCs. However, emission rates of isoprene are higher in EU buildings. This could be due the presence of other significant sources in EU buildings. Possible sources include latex paints, synthetic rubbers, adhesives, tapes and wire coatings. However, it is highly possible that the main source of isoprene is from ETS.

CONCLUSION AND IMPLICATIONS

This study highlights the presence of dominant source/s of several VOCs and their geographical similarities and differences within EU and Singapore buildings. Concentrations and emission rates of 2-methylpentane and heptane are higher in EU buildings as compared to the ones in Singapore. Also, lower alkanes are observed to be higher in levels in EU buildings as opposed to the higher alkanes which are more dominant in Singapore buildings with statistical differences approaching significance. The geographical difference in emission rates revealed higher emission rates in EU buildings for three VOCs as opposed to two for differences in concentrations. This could be due to the significantly higher ACH in EU buildings as compared to that in Singapore. Benzene and toluene concentrations and emission rates are significantly higher in ETS-free Singapore buildings. There are no differences in VOCs (TCA and limonene) related to occupant suggesting similar use of consumer products. However, differences in emission rates of limonene approach significance which maybe related to wood products commonly used in European buildings or ETS. In EU buildings, higher emission rates for isoprene could be due to ETS instead of bioeffluents emissions while for 2-methylpentane, it is due to solvent use.

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