Impact of ventilation and recirculation rates on exposure to and intake of ozone and its initiated chemistry products: Mass balance model evaluation

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ABSTRACT

A mass balance model is used to examine the impact of two ventilation (1 /h and 2 /h) and recirculation (7 /h and 14 /h) rates on concentrations, exposure to and intake of ozone (of outdoor origin) and secondary organic aerosols (SOA) derived from the ozone initiated chemistry in indoor environment. Measured data from several experimental studies conducted by the authors in a 236m^3 field environmental chamber (FEC) configured to simulate an office are used for the mass balance model evaluations. At steady state, increase in ventilation rate increases exposure to and intake of indoor ozone, but reduces exposure to and intake of SOA. Increase in recirculation rate reduces exposure to and intake of ozone and SOA. Increase in outdoor ozone concentration increases exposure to and intake of ozone and SOA. As expected, indoor ozone and SOA concentrations are lower for human occupancy scenario than non-human occupancy scenario. Interestingly, human sink of ozone and SOA is much lower at higher recirculation rate than at lower recirculation rate - this is a new finding that has not been reported in the literature.

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KEYWORDS

Ventilation rate; Recirculation rate; Ozone; Indoor chemistry; Exposure and intake

1 INTRODUCTION

A foremost energy challenge in the 21st century is influencing how buildings are being designed to reduce exchange between outdoor and indoor air (Mudarri, 2010). The rate at which outdoor and indoor air are deliberately exchanged (ventilation) could influence building occupants' exposure to and intake of pollutants of outdoor and indoor origins. In this paper, pollutant of interest is ozone. Ozone is chosen for three reasons.

First, the outdoors is a major source of ozone in indoor environment (Weschler, 2000). Second, increase in exposure to and intake of ozone has been associated with increase in hospital visits, morbidity and mortality rates (Bell et al. 2004). Third, ozone is a powerful oxidizing agent and products of its initiated chemistry could be more harmful than ozone alone (Rohr, 2013).

Due to energy concerns, recirculation of a larger proportion of indoor air has become a normal practice in air-conditioned buildings located in regions with warm and humid outdoor air. The effects of ventilation and recirculation rates on ozone and its initiated chemistry products have been reported in the literature (Zuraimi et al. 2007; Fadeyi et al. 2009; Fadeyi, 2015). These studies were done with no humans present in the indoor environment. However, in reality humans will be present in buildings – and this should be the primary reason why these studies were conducted in the first place. Furthermore, evidence have shown that when human are present in the indoor environment, they change the dynamics and concentrations of ozone and its initiated chemistry products (see references in Weschler, 2015).

Fadeyi et al (2013) examined the impact of human presence on ozone and secondary organic aerosols (SOA) derived from ozone initiated chemistry products, when the air-conditioned system recirculates a large proportion of indoor air, and ventilation transports outdoor air, containing ozone, to indoor environment. The key finding from their study is that human presence would reduce concentrations of indoor ozone and SOA. Fadeyi et al. (2013) and previous studies did not specifically address exposure to and intake of these pollutants – ozone and SOA. There is also no evidence on how changes in ventilation and recirculation rates would influence the dynamics and concentrations of ozone and SOA, exposure and intake when human are present in indoor environment for considerable amount of time.

To bridge this gap in knowledge, a mass-balance model evaluation is used to examine how ventilation and recirculation rates would influence concentrations, exposure to and intake of ozone and SOA derived from ozone initiated chemistry. Additionally, impact of increased outdoor ozone concentration on exposure to and intake of ozone and SOA is examined. This study gives indication of how changes in ventilation and recirculation rates could influence concentrations, exposure to and intake of pollutants of outdoor and indoor origins.

2 METHODS

The mass balance model adopted in this study used measured data from experimental studies conducted by the authors in a 236m^3 field environmental chamber (FEC) configured to simulate an office. The air system of the FEC operates under recirculation mode with a recirculation loop of ~30m^3. Details of the experiments and FEC can be found in Fadeyi et al. (2009, 2013) and Zuraimi et al (2007). The mass balance model (see Equations 1and 2) used in this study has been validated with experiments conducted by Fadeyi et al. (2009).

In this study, the mass balance model evaluation is used to evaluate a hypothetical 3-h exposure and intake period – time from when steady state has been achieved in the FEC. We did not account for exposure and intake during transient reactions – period leading to the steady state – that occurred when occupants just entered the FEC. Specifically, the mass balance model is used to examine the impact of two ventilation (1 /h and 2 /h) and two recirculation (7 /h and 14 /h) rates on concentrations, exposure to and intake of ozone (of outdoor origin) and SOA derived from ozone reacting with limonene emitted in the FEC at a constant emission rate of 180mg/h throughout the steady state 3-h occupancy of the FEC. The impact of increase in ventilation rate from 1 /h to 2 /h is calculated for doubling the “emission rate of ozone” – with assumed outdoor concentration of 157µg/m^3 (80ppb) – transported from outdoor to indoor. Ozone emission “E_{O3}” rates into the FEC are calculated to be 41.8 and 83.5mg/h at ventilation rates of 1 /h and 2 /h, respectively.

$$[C_{O3}]_{ss} = \frac{(\eta_v \lambda_v + \rho \lambda_L)\, C_{out} + (E_{O3}/V)}{(\lambda_v + \lambda_L + k_{sr,O3} + k_{O3,lim}[lim]_{ss})} \tag{1}$$

$$[C_{SOA}]_{ss} = \frac{\{\acute{y}_{SOA}\%\ k_{O3,lim}[lim]_{ss}\ [C_{O3}]_{ss})/V\}}{\{\lambda_v + \lambda_L + f\,(\lambda_v + \lambda_{recirc}) + k_{sr,SOA}\}} \tag{2}$$

“$[C_{O3}]_{ss}$” is the concentration of ozone in the FEC at steady state. “$(\eta_v\lambda_v)\ C_{out}$” is the contribution of outdoor environment to ozone in the FEC through fraction (η_v=1, i.e. assuming 100% penetration of outdoor ozone to indoor as it is introduced together with ventilation) of outdoor ozone (C_{out} – assumed to be 157µg/m) that enters the FEC through dedicated outdoor inlet meant for outdoor air.

266m^3 is the volume “V” of the system – summation of FEC and recirculation loop volumes. The values, based on experimental data from Fadeyi et al. (2009), used for “$k_{sr,O3}$” – the rate at which ozone is removed by indoor surfaces – are 2.96 /h and 6.73 /h for ventilation rates of 1 /h and 2 /h, respectively after adjusting for assumed no leakages in the system – i.e. $\lambda_L = 0$, thus penetration factor “ρ” is also “0”.

$k_{O3,lim}[lim]_{ss}$” is the rate at which ozone is removed from the FEC by limonene through chemical reactions; 0.018 $ppb^{-1}h^{-1}$ is used for “$k_{O3,lim}$” – the second rate constant for the reactions between ozone and limonene (Atkinson et al. 1990);“$[lim]_{ss}$” is the concentration of limonene at steady state.

“$[C_{SOA}]_{ss}$” is the concentration of SOA formed in the FEC at steady state. “$\{\acute{y}_{SOA}\%\ k_{O3,lim}[lim]_{ss}\ [C_{O3}]_{ss})$” is the rate at which SOA is generated in the FEC, while $\acute{y}_{SOA}$ is the yield of formation of SOA. 0.16 and 0.14 are used as “$\acute{y}_{SOA}$” values for ventilation rate (λ_v) of 1 /h and 2 /h, respectively (Youssefi and Waring, 2014). 0.39 is used for new filter filtration efficiency “f” (Fadeyi et al. 2009). “$k_{sr,SOA}$” is the rate at which SOA is removed by indoor surfaces. 0.5 /h and 1/h are “$k_{sr,SOA}$” values at recirculation rate “λ_{recirc}” of 7 /h and 14 /h (Zuraimi et al. 2007).

Equations 3 and 4 show parameters that would determine concentrations of ozone and SOA in the FEC, respectively for a scenario when the FEC is occupied by 18 occupants – typical capacity of the FEC. “$k_{sr,O3\_human}$”, rate at which ozone is removed by human surface is calculated, from experimental data, to be ~2 /h for both recirculation rates of 7/h (Fadeyi et al. 2013). The same value – ~2 /h – is hypothetically used for recirculation rate of 14 /h. “k_{O3_inhal}” – the rate at which ozone is removed by human through inhalation – is assumed to be negligible.

“k_{sr,SOA_human}” is the rate at which SOA is removed by human – 0.1 /h is used for recirculation rate of 7 /h (Fadeyi et al. 2013). 0.2 /h (value for “k_{sr,SOA_human}”) is hypothetically used for recirculation rate of 14 /h. 0.04 /h is used for “k_{SOA_inhal}” – the rate at which SOA is removed by human through inhalation (Fadeyi et al. 2013). Other parameters have been defined earlier. 3-h ozone and SOA exposure are calculated by multiplying calculated ozone and SOA concentrations, using Equation 3 and 4, values by 3 – hypothetical 3-h exposure at steady state and intake period.

$$[C_{O3}]_{ss} = \frac{(\eta_v\lambda_v + \rho\lambda_L)\ C_{out} + (E_{O3}/V)}{(\lambda_v + \lambda_L + k_{sr,O3} + k_{sr,O3\_human} + k_{inhal} + k_{O3,lim}[lim]_{ss})} \tag{3}$$

$$[C_{SOA}]_{ss} = \frac{\{\acute{y}_{SOA}\%\ k_{O3,lim}[lim]_{ss}\ [C_{O3}]_{ss})/V\}}{\{\lambda_v + \lambda_L + f\,(\lambda_v + \lambda_{recirc}) + k_{sr,SOA} + k_{sr,\ SOA\_human} + k_{SOA\_inhal}\}} \tag{4}$$

The 3-h ozone and SOA intakes by the occupants are calculated using Equations 5 and 6 – written based on knowledge gained from Weschler (2006). Scenario where adults are the exposed occupants was examined. Assuming occupants performed sedentary activities throughout their occupancy of the FEC, their breathing rate (BR_{indoor}) was assumed to be $0.54 m^3/h$ (Weschler, 2006).

$$\text{3-h } [C_{O3}]_{ss} \text{ intake} = 3\text{x } [C_{O3}]_{ss} \text{ x } BR_{indoor} \quad (5)$$

$$\text{3-h } [C_{SOA}]_{ss} \text{ intake} = 3\text{x } [C_{SOA}]_{ss} \text{ x } BR_{indoor} \quad (6)$$

3 RESULTS

Increase in ventilation rate – from 1 /h to 2 /h, when source of ozone is from outdoors, increases indoor ozone concentration (see Figure 1). This impact of increased ventilation rate on indoor ozone is reduced by increasing recirculation rate from 7 /h to 14 /h. At 7 /h, the differences in ozone concentrations are calculated to be 6.7 and 5.3µg/m^3 for "human" and "no human" status, respectively. At 14 /h, the differences in ozone concentrations are calculated to be 4.5µg/m^3 for both occupancy statuses.

Increase in recirculation rate – from 7 /h to 14 /h – reduces indoor ozone concentrations. This effect is more evident when the FEC is not occupied. When FEC is occupied, increased recirculation rate reduces ozone concentrations by 10.3 and 12.5µg/m^3 for ventilation rates of 1 /h and 2 /h, respectively. When FEC is not occupied, increased recirculation rate reduces ozone concentrations by 15.9 and 16.7µg/m^3 for ventilation rates of 1 /h and 2 /h, respectively.

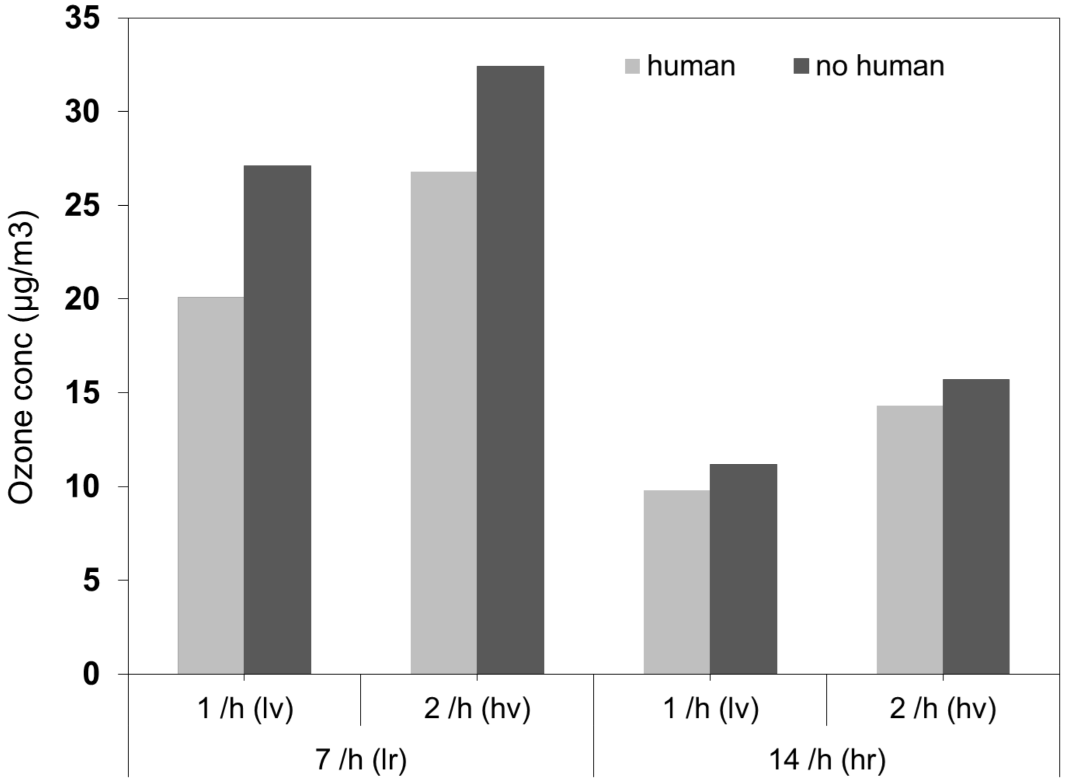

Figure 1: Impact of ventilation and recirculation rates on ozone concentration during human and non-human presence in the FEC.

As evident in Figure 1, additional sink provided by human presence causes indoor ozone concentrations to be lower than when it is not occupied ("no human"), irrespective of ventilation and recirculation rates. Interestingly, the sink effect diminishes when recirculation rate is increased from 7 /h to 14 /h. At recirculation rate of 7 /h, the decrease is calculated to be 7 and 5.6µg/m^3, for ventilation rates of 1 /h and 2 /h, respectively. At recirculation rate of 14 /h, the decrease is calculated to be 1.4µg/m^3 for both ventilation rates.

Figure 2 shows impact of ventilation and recirculation rates on SOA concentrations during human and non-human presence in the FEC. Increase in ventilation rate – from 1 /h to 2 /h – reduces SOA concentrations. This impact of increased ventilation rate on indoor SOA concentrations is reduced by increasing recirculation rate from 7 /h to 14 /h. At 7 /h, the differences in SOA concentrations are calculated to be 2.5 and 3.6µg/m^3 for "human" and "no human" status, respectively. At 14 /h, the differences in ozone concentrations are calculated to be 0.6 and 0.8 µg/m^3 for "human" and "no human" status, respectively.

Increase in recirculation rate – from 7 /h to 14 /h – reduces SOA concentrations (see Figure 2). This effect is more evident when the FEC is not occupied for each of the ventilation rate. At 1 /h, increased recirculation rate reduces SOA concentrations by 3.4 and 4.7µg/m^3 for "human" and "no human", respectively. At 2 /h, increased recirculation rate reduces SOA concentrations by 1.5 and 1.9µg/m^3 for "human" and "no human", respectively.

Lower SOA concentrations occur when the FEC is occupied than when it is not occupied, irrespective of ventilation and recirculation rates (see Figure 2). Like in the case of ozone, sink effect due to human presence diminishes when recirculation rate is increased from 7 /h to 14 /h. At recirculation rate of 7 /h, the decrease is calculated to be 1.6 and 0.5µg/m^3 for ventilation rates of 1 /h and 2 /h, respectively. At recirculation rate of 14 /h, the decrease is calculated to be 0.3 and 0.1µg/m^3 for ventilation rates of 1 /h and 2 /h, respectively.

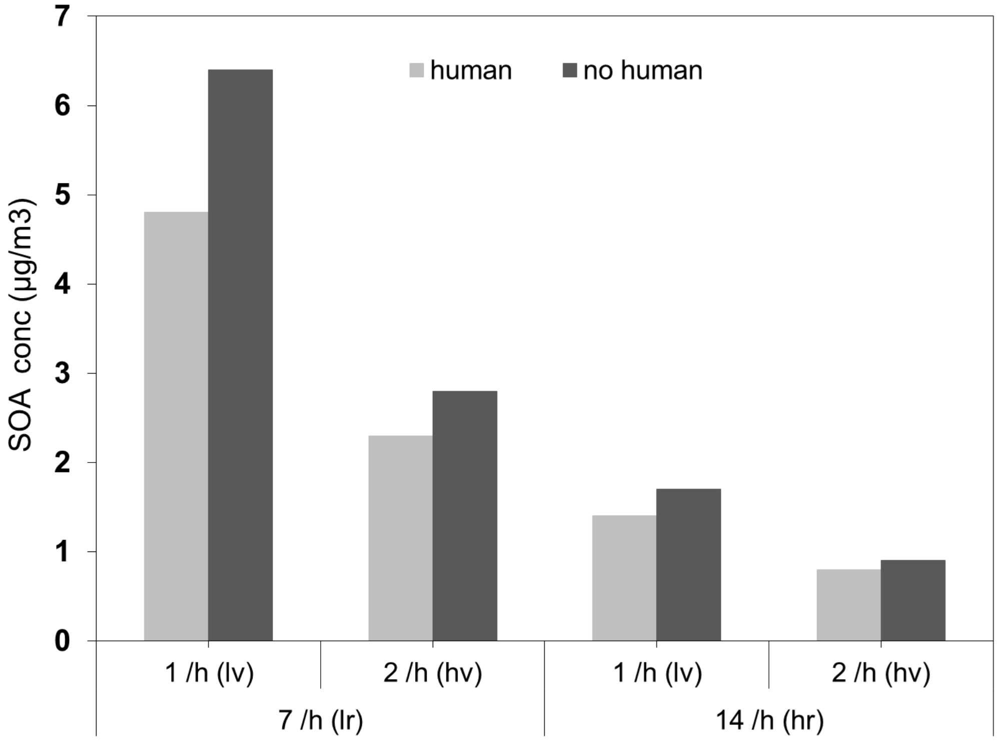

Figure 2: Impact of ventilation and recirculation rates on SOA concentrations during human and non-human presence in the FEC.

Figure 3 shows 3-h exposure to and intake of ozone at different ventilation and recirculation rates. Increase in ventilation rate – from 1 /h to 2 /h – increases building occupants' 3-h ozone exposures and intakes. The effect of increased ventilation rate causing higher exposure and intake is more evident at lower recirculation rate. Increases in ozone exposures are calculated to be 20.1 and 13.4µg/m^{3}h at recirculation rates of 7 /h and 14 /h, respectively. Increases in ozone intakes are calculated to be 10.9 and 7.2µg at recirculation rates of 7 /h and 14 /h, respectively.

Increase in recirculation rate – from 7 /h to 14 /h – reduces occupants' 3-h ozone exposures and intakes. The effect of increased recirculation rate causing lower ozone exposure and intake is more evident at higher ventilation rate when more outdoor ozone is transported indoor. Reductions in ozone exposures, caused by increasing recirculation rate, are calculated to be 31 and 37.7µg/m^{3}h at ventilation rates of 1 /h and 2 /h, respectively. The reductions in ozone intakes, caused by increasing recirculation rate, are calculated to be 16.7 and 20.4µg at ventilation rates of 1 /h and 2 /h, respectively.

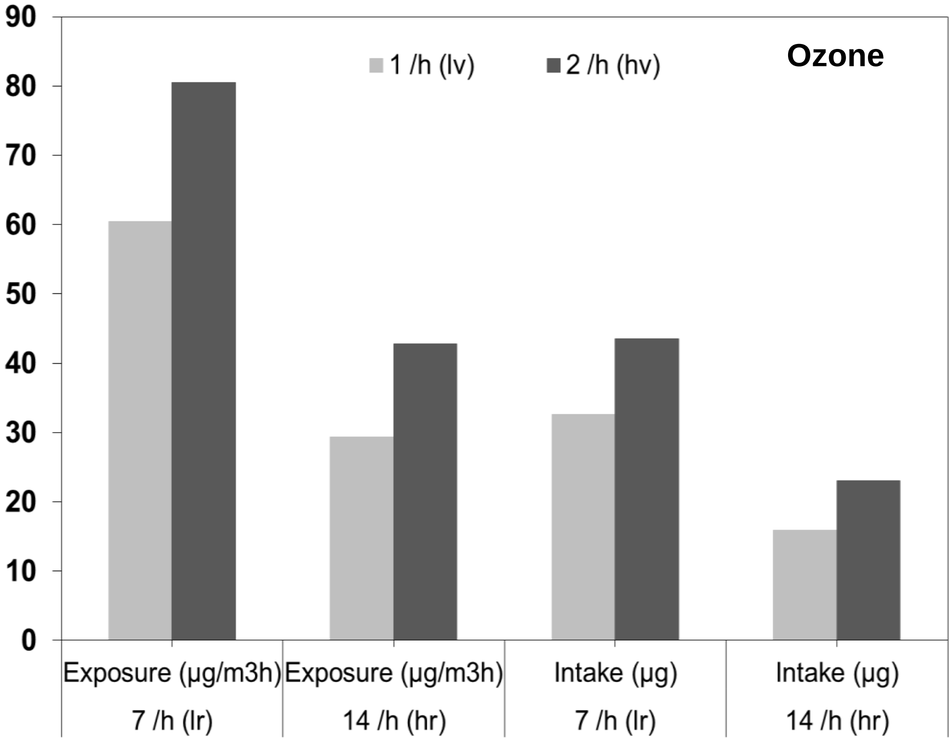

Figure 3: 3-h exposure to and intake of ozone at different ventilation and recirculation rates

Figure 4 shows 3-h exposure to and intake of SOA at different ventilation and recirculation rates. Increase in ventilation rate – from 1 /h to 2 /h – reduces occupants' 3-h SOA exposures and intakes. The effect of increased ventilation rate causing lower SOA exposure and intake is more evident at lower recirculation rate. Reductions in SOA exposures, caused by increasing ventilation rate, are calculated to be 7.6 and 1.9µg/m^{3}h at recirculation rates of 7 /h and 14 /h, respectively. Reductions in SOA intakes, caused by increasing ventilation rate, are calculated to be 4.1 and 1µg at recirculation rates of 7 /h and 14 /h, respectively.

Increase in recirculation rate – from 7 /h to 14 /h – reduces building occupants' 3-h SOA exposures and intakes. The effect of increased recirculation rate causing lower SOA exposure and intake is more evident at lower ventilation rate. Reductions in SOA exposures, caused by increasing recirculation rate, are calculated to be 10.2 and 4.5µg/m^{3}h at ventilation rates of 1 /h and 2 /h, respectively. Reductions in SOA intakes, caused by increasing recirculation rate, are calculated to be 5.5 and 2.4µg at recirculation rates of 7 /h and 14 /h, respectively.

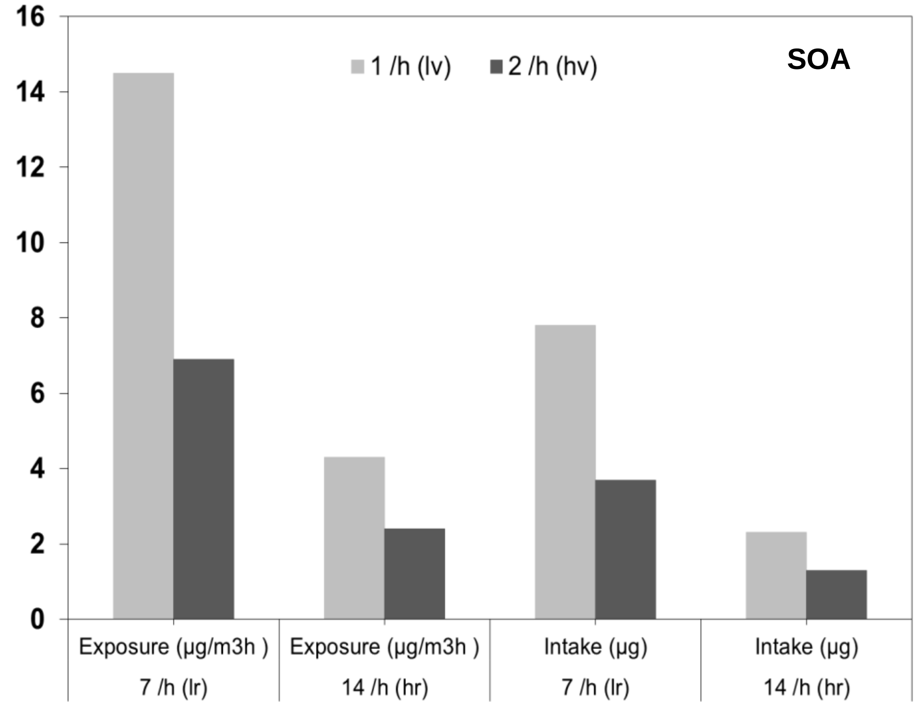

Figure 4: 3-h exposure to and intake of SOA at different ventilation and recirculation rates

Figure 5 shows exposure to and intake of ozone and SOA at different outdoor ozone concentrations. Increase in outdoor ozone concentration increases exposure to and intake of ozone and SOA in the FEC.

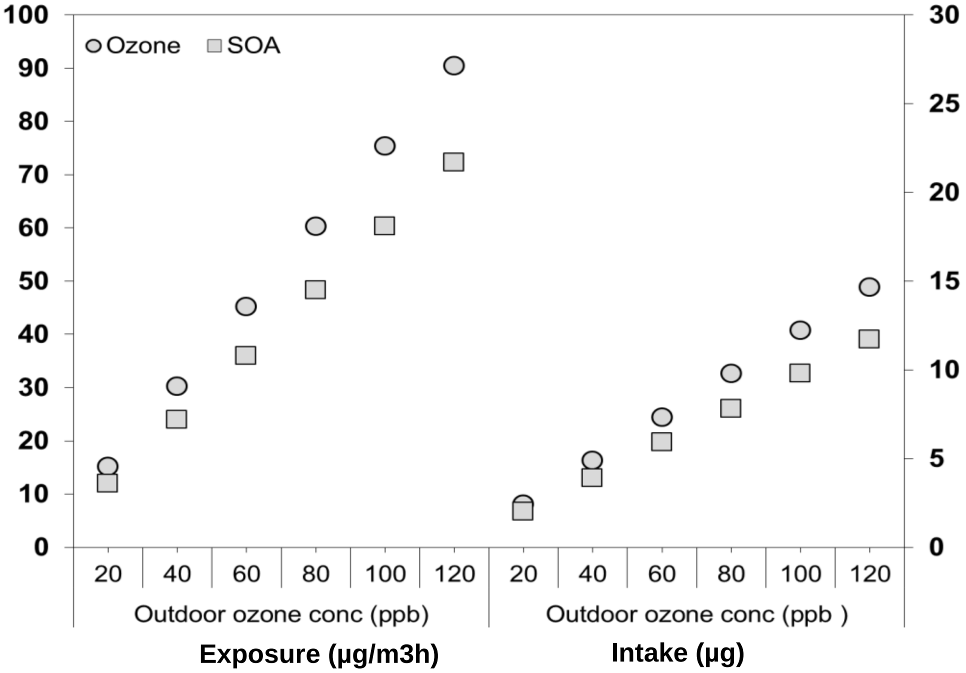

Figure 5: Exposure to and intake of ozone and SOA at different outdoor ozone concentrations

4 DISCUSSION

4.1 Impact of ventilation and recirculation rates and human presence

With ozone coming from outdoors, increase in ventilation rate will increase indoor ozone concentration because of increased outdoor to indoor transport of ozone. However, increase in ventilation rate did not increase SOA concentration because SOA is a pollutant of indoor source. In this case, for every increase in ventilation rate, dilution of SOA will increase. Additionally, the resident time – available time – for ozone to react with limonene to generate SOA is lower at higher ventilation rate (Weschler and Shield 2013). This phenomenon leads to lesser SOA formation at higher ventilation rate than at lower ventilation rate which has higher resident time. Boundary layer of surfaces in the FEC and air system become thinner as air moves at a much faster rate when recirculation rate is set higher (Zuraimi et al. 2007). The thinner nature of the surfaces enhance surface removal rate of ozone and SOA from the gas phase. This explains why higher recirculation rate causes lower indoor ozone and SOA concentrations.

When humans are present in the FEC, it means additional sink for ozone and SOA are introduced (Fadeyi et al. 2013). This explains why ozone and SOA concentrations are lower when FEC is occupied. At higher recirculation, the amount of ozone and SOA available at the gas phase is much lower, due to much higher surface removal of ozone and SOA by material surfaces in the FEC and air system. Thus, the ozone and SOA sink effect due to human presence, although still evident, diminishes – this observation is more evident in the case of ozone which is more prone to human sink effect than SOA. To the authors´ knowledge this is a new finding and it is being reported for the first time.

Decrease in ventilation rate increases SOA exposure and intake. However, by moving indoor air in the FEC and air system at a much faster rate – i.e. at higher recirculation rate – the effect of lower resident time causing more SOA will be diminished. This is due to increased surface removal of ozone caused by higher recirculation rate. In this study, ozone is assumed to be introduced from the outdoors. Thus, increased ventilation rate increases ozone exposure and intake. However, increased ozone exposure and intake caused by increased ventilation rate diminishes by moving air at a much faster rate. It is important to note that increased ventilation rate still increases outdoor to indoor transport of ozone, but the effect of this transport on increased indoor ozone concentration at steady state reduces because of higher surface removal of ozone at the gas phase by higher recirculation rate. These findings have implication on energy savings. Setting ventilation rate low especially when outdoor air poses threat of introducing pollutant(s) to indoor environment can be used to conserve energy, while using higher recirculation rate which has relatively lower energy consumption when compared to increasing ventilation rate to reduce indoor pollutants (Fadeyi and Tham 2008).

The difference between SOA exposure and intake when air is moved at a faster rate (14 /h) – and lower rate (7 /h) – is much higher than the difference experienced in the case of ozone. This is because higher recirculation rate and ventilation reduces SOA concentration; however, ventilation introduces more ozone while higher recirculation reduces ozone. Increase in outdoor ozone concentration, when there is air exchange between outdoor and indoor air, increases the vulnerability of indoor occupants to higher ozone and SOA exposure and intake. This finding poses a concern for cities and regions experiencing high outdoor pollutants, not necessary ozone alone, caused by natural and human factors. Such occurrence also poses threats to gaining benefits inherent in the usage of ventilation to reduce indoor pollutants.

4.2 Uncertainly in mass-balance modeling

Only few ozone data points were used for calculations of ozone surface removal rate by the system “$k_{sr}O_3$” at the higher recirculation rate with filter placed in the AHU. This is because ozone decayed at a faster rate. This limitation caused calculated “ksrO$_3$” values to be less accurate. “$k_{sr}O_3$” values are more accurate for lower recirculation rate because ozone decayed at a slower rate and more data were used for the calculation.

For simplification purpose, the mass-balance modelling of SOA particles was based on a single particle size (~100 nm) to represent all SOA particles. Computation of SOA particles by properly accounting for all SOA particles measured during the experiments would produce more accurate results. Changes in outdoor particle concentrations during experiments reduced the accuracy of measured data used for this mass balance modelling.

Due to instrumentation error, limonene concentration was computed and not measured directly by Fadeyi et al. (2009). All hypothetically assumed values may be less accurate. In this study, the system is assumed to have no leakage. However, leakage is a common occurrence in buildings. When leakage occurs, it may change the impact of outdoor air change rate on steady state indoor ozone concentration (Fadeyi et al. 2009). Finally, exposure and intake analysis reported in this paper are based on steady state period. The findings from this analysis may not be applicable for non-steady state period.

5 CONCLUSIONS

How would changes in ventilation and recirculation rates influence building occupants’ exposure to and intake of ozone and by-products of ozone initiated chemistry? A mass balance model evaluation, based on several experiments conducted by the authors, was used in an attempt to answer this question. The following are the main conclusions from this exercise.

- Increase in ventilation rate, with ozone originating from outdoor, increases indoor ozone concentration, exposure and intake, but reduces SOA concentration, exposure and intake.
- Increase in recirculation rate reduces ozone and SOA concentrations, exposure and intake.
- Indoor ozone and SOA concentrations are lower for human occupancy scenario than non-human occupancy scenario.
- Human sink of ozone and SOA is much lower at higher recirculation rate than at lower recirculation rate. This finding has never been reported before in the literature.
- Increase in outdoor ozone concentration increases outdoor to indoor transport of ozone, thereby causing higher exposure to and intake of ozone and SOA.

6 ACKNOWLEDGEMENTS

The financial assistance from the National University of Singapore is gratefully acknowledged through grants R296-000-117-112. Dr. Moshood Olawale Fadeyi participation in this research is partly funded by the Republic of Singapore’s National Research Foundation through a grant to the Berkeley Education Alliance for Research in Singapore (BEARS) for the Singapore-Berkeley Building Efficiency and Sustainability in the Tropics (SinBerBEST) Program. The contributions of Prof. Charles J. Weschler (CJW), Dr. Zuraimi M. Sultan, and Miss Wu W. Yi to experimental studies used for the mass balance model evaluation are gratefully acknowledged. We thank CJW for his advice in the construction of the mass balance model.

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