

Technical note

Barriers and opportunities for passive removal of indoor ozone

Elliott T. Gall, Richard L. Corsi, Jeffrey A. Siegel*

University of Texas at Austin, Austin, TX, USA

ARTICLE INFO

Article history:

Received 14 November 2010

Received in revised form

9 March 2011

Accepted 11 March 2011

Keywords:

Indoor air quality

Passive pollutant removal

Low-energy air cleaning

ABSTRACT

This paper presents a Monte Carlo simulation to assess passive removal materials (PRMs) that remove ozone with no additional energy input and minimal byproduct formation. Distributions for air exchange rate in a subset of homes in Houston, Texas, were taken from the literature and combined with background ozone removal rates in typical houses and previous experimentally determined ozone deposition velocities to activated carbon cloth and gypsum wallboard PRMs. The median ratio of indoor to outdoor ozone was predicted to be 0.16 for homes with no PRMs installed and ranged from 0.047 to 0.12 for homes with PRMs. Median values of ozone removal effectiveness in these homes ranged from 22% to 68% for the conditions investigated. Achieving an ozone removal effectiveness above 50% in half of the homes would require installing a large area of PRMs and providing enhanced air speed to transport pollutants to PRM surfaces. Challenges associated with achieving this removal include optimizing indoor transport and aesthetic implications of large surface areas of PRM materials.

© 2011 Elsevier Ltd. All rights reserved.

1. Introduction

As building energy use decreases with measures aimed towards increasing building energy efficiency, sustainable building protocols incorporate designs to recirculate a greater fraction of indoor air. This is because ventilation with outdoor air accounts for 36% of space conditioning energy, or 13% of total building energy, on average (Liddament and Orme., 1998; DOE, 2006). Two major drawbacks to the energy efficiency gains realized from reducing ventilation are: 1) the increased potential for exposure to a wide variety of pollutants of indoor origin, and 2) the increased potential for indoor reactions, often driven by ozone, that can result in harmful byproducts (Weschler, 2006). Meeting demands for healthy indoor environments in conjunction with energy efficiency goals will require new approaches to indoor air cleaning.

The improvement of indoor air generally involves systems that require electrical energy, such as heating, ventilation and air conditioning (HVAC) filtration, increased ventilation for dilution of indoor pollutants, or stand-alone air purifiers. In this paper, we explore the concept of passive removal materials (PRMs), defined as materials that result in the removal of indoor pollutants without generating harmful byproducts and requiring no additional energy input beyond normal building operation (Kunkel et al., 2010; Sekine and Nishimura, 2001). We specifically focus on ozone removal because increases in ozone exposure are associated with increased

mortality and asthma morbidity (Bell et al., 2005). Ozone is also highly reactive, resulting in reaction byproducts that are potentially more damaging than ozone itself (Weschler, 2006).

We utilize Monte Carlo simulation to characterize the opportunity for indoor ozone removal utilizing PRMs in homes in Houston, TX. Houston is an ideal setting for the modeling of PRMs due to its history of ozone exceedance and the availability of data for 164 homes from the Relationship of Indoor, Outdoor and Personal Air (RIOPA) investigation (Weisel et al., 2005). Using findings from this subset of homes, the opportunity for ozone removal by PRMs is discussed in terms of three categories of potential barriers to implementation: 1) transport of ozone to PRMs, 2) PRM surface area, and 3) potential for byproduct formation from ozone-PRM interactions.

2. Mass balance and parameters

The dominant source of ozone in buildings is outdoor air and indoor ozone concentrations are often expressed as a fraction of the outdoor concentration. Equation (1) is a time-averaged solution for indoor/outdoor ozone ratio for a hypothetical well-mixed home:

$$\frac{C}{C_o} = \frac{1}{1 + v_{d,BG} \frac{A_{BG}}{\lambda V} + v_{d,PRM} \frac{A_{PRM}}{\lambda V}} \quad (1)$$

Where C is the ozone concentration in an indoor environment (ppb), C_o is the outdoor concentration of ozone entering the indoor

* Corresponding author.

E-mail address: jasiegel@mail.utexas.edu (J.A. Siegel).

environment, v_d is the deposition velocity to indoor surfaces (m h^{-1}), A is the horizontally projected surface area of indoor surfaces (m^2), λ is the air exchange rate (h^{-1}), V is the volume of the indoor environment (m^3), the *BG* subscript indicates that parameters refer to background surfaces present in indoor environments, and the *PRM* subscript indicates that parameters refer to surfaces associated with PRMs. The central metric, effectiveness, is calculated by dividing the indoor/outdoor ratios of ozone concentration with PRMs present (Equation (1) above) and without the presence of PRMs ($A_{PRM} = 0$ in Equation (1)) and subtracting from unity (Miller-Leiden et al., 1996; Kunkel et al., 2010). Effectiveness has a value of unity when the PRM removes all indoor ozone and a value of zero when the PRM has no impact on indoor ozone. Removal effectiveness was estimated for the hypothetical presence of two materials (PRMs): gypsum wallboard (GWB) and activated carbon cloth (ACC).

Table 1 summarizes the parameters used in the Monte Carlo simulation based on Equation (1). A normal statistical distribution was generated for the background decay rate of ozone in homes, with descriptive parameters (arithmetic mean = 2.8 h^{-1} , arithmetic standard deviation = 1.3 h^{-1}) taken from a study of ozone decay rates in residences (Lee et al., 1999). Yamamoto et al. (2010) reported a median air exchange rate of 0.47 h^{-1} for Texas homes in the RIOPA study. For this study we use a geometric mean of 0.5 h^{-1} with a geometric standard deviation of 2.13 h^{-1} (Weisel et al., 2005).

The surface area of installed PRMs and the deposition velocity to PRMs were simultaneously varied to high and low conditions for each PRM. “High” and “Low” designations following PRM material names refer to model parameter inputs, e.g., *GWB-High* refers to gypsum wallboard installed over a large surface area and utilizing high deposition velocity. The large surface area to volume ratio (0.3 m^{-1}) was selected to approximate a PRM installation equivalent to the projected area of a home’s ceiling. The low surface area of PRM to volume ratio (0.075 m^{-1}) was selected to approximate a PRM installation equivalent to covering the walls of one bedroom. Ozone deposition velocities to PRMs were taken as constant values based on experimental results at low and high air speeds reported previously (Kunkel et al., 2010). The number of iterations conducted in the Monte Carlo simulation was increased from 1000 in increments of 1000 until differences in median effectiveness values were less than 1% after increasing simulation iterations (occurring at 10,000 iterations).

3. Results and discussion

3.1. Ozone removal effectiveness

Cumulative distribution functions (CDFs) of effectiveness for the four scenarios considered are shown in Fig. 1. The median and mean

Table 1
Summary of parameters.

Variable	Value	Notes
Background ozone removal	2.8 h^{-1} (1.3 h^{-1})	AM (ASD) Lee et al. (1999)
Air exchange rate	0.5 h^{-1} (2.13)	GM (GSD) Weisel et al. (2005)
Surface area to volume ratio	0.075 m^{-1}	Low condition. Assumed
	0.3 m^{-1}	High condition. Assumed
Deposition velocity ^a	13 m h^{-1}	GWB-low condition. Kunkel et al. (2010)
	13.2 m h^{-1}	GWB-high condition. Ibid.
	20.8 m h^{-1}	ACC-low condition. Ibid.
	23.5 m h^{-1}	ACC-high condition. Ibid.

AM = arithmetic mean, ASD = arithmetic standard deviation, GM = geometric mean, GSD = geometric standard deviation, *GWB* = gypsum wallboard, *ACC* = activated carbon cloth.

^a Deposition velocity for low condition taken at an indoor air speed of 0.42 m s^{-1} . High conditions taken at an indoor air speed of 0.75 m s^{-1} . (Kunkel et al., 2010).

background indoor/outdoor ozone ratios estimated in this study are 0.16 and 0.17, respectively. This background condition generally agrees with experimentally determined indoor/outdoor ratios summarized by Weschler (2000). The presence of PRMs in RIOPA homes reduces the median indoor/outdoor ozone ratio to 0.05, 0.07, 0.10, and 0.12 for *ACC-High*, *GWB-High*, *ACC-Low* and *GWB-Low* conditions, respectively. Combinations of low deposition velocities and high surface areas were modeled in the Monte Carlo simulation, however, at the elevated experimental air speeds performed by Kunkel et al. (2010) differences in deposition velocity to *ACC* and *GWB* are moderate relative to the modeled differences in surface area. Therefore, only combinations of high surface area, high deposition velocity and low surface area, low deposition velocity are shown. Combinations of high surface area and low deposition velocity result in median values of effectiveness of 67% and 54% in the modeled homes for *ACC* and *GWB*, respectively. Combinations of low surface area and high deposition velocity result in median values of effectiveness of 35% and 24% in the modeled homes for *ACC* and *GWB*, respectively.

An ozone removal effectiveness of 50% represents a value where the ozone removal contribution from PRMs is equivalent to other removal mechanisms considered (ventilation and background removal), and is a proposed minimum threshold for air cleaning devices (Shaughnessy and Sextro, 2006). Fig. 1 shows that 10% and 20% of RIOPA homes realize this threshold for *GWB-Low* and *ACC-Low* conditions, respectively. However, the *GWB-High* and *ACC-High* conditions surpass the 50% effectiveness threshold in 65% and 100% of RIOPA homes, respectively. The recommended effectiveness of 80% noted by Shaughnessy and Sextro (2006) is achieved by no homes under Low conditions, and approximately 10% and 25% of RIOPA homes under *GWB-High* and *ACC-High* conditions, respectively. Increasing PRM surface area beyond the High conditions considered here would increase the number of homes achieving this recommended effectiveness level. PRMs, however, can largely achieve minimum effectiveness values and therefore should be considered as a credible option for indoor air cleaning, potentially in combination with other ozone control strategies. Mandates limiting the energy use of buildings lend further support to pursuing implementation of PRMs in buildings.

3.2. Enhancing transport to PRMs

By definition, PRMs should be relatively reactive with ozone and, as such, removal of ozone will be largely dependent on

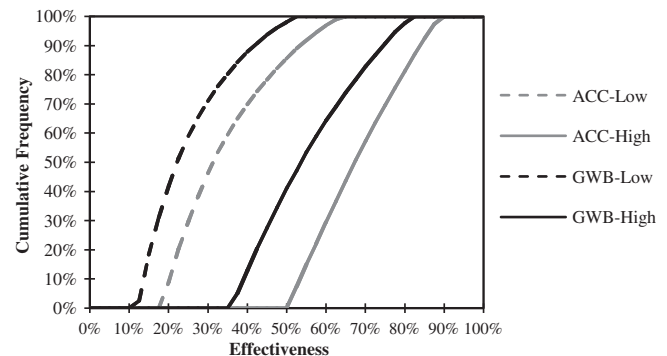


Fig. 1. Mass balance cumulative distribution functions illustrating effectiveness of PRMs in Houston homes. *ACC* = activated carbon cloth. *GWB* = gypsum wallboard. Low condition refers to deposition velocities of 13 m h^{-1} and 20.8 m h^{-1} for gypsum wallboard and activated carbon cloth, respectively, and a surface area to volume ratio of 0.075 m^{-1} . High condition refers to deposition velocities of 13.2 m h^{-1} and 23.5 m h^{-1} for gypsum wallboard and activated carbon cloth, respectively, and a surface area to volume ratio of 0.3 m^{-1} .

transport to the PRM. Kunkel et al. (2010) reported promising experimental results that illustrate the potential of enhanced pollutant transport to PRMs at elevated air speeds. These experiments utilize a small area ($<1 \text{ m}^2$) of activated carbon cloth placed around ceiling fan blades in a living room, resulting in an increase in ozone decay rate of 33% above background (effectiveness = 44%, assuming the mean air exchange from the RIOPA study). Elevated air speed in the region of PRMs is desirable for enhanced mass transport. The range of air speeds corresponding with deposition velocities to materials used in this study ranged from 0.42 to 0.75 m s^{-1} , measured 10 cm away from the material during previous experiments in a controlled test house (Kunkel et al., 2010). These air speeds are higher than what is found in bulk air of typical indoor environments, which generally range from 0.05 to 0.15 m s^{-1} (Matthews et al., 1989). Model simulations utilizing this typical range of indoor air speeds would result in decreased effectiveness compared to results presented in Fig. 1. In residential buildings, air speed above typical ranges could correspond with southern facing walls with convective currents or areas downstream of ceiling and floor fans. Regions downstream of HVAC supply diffusers provide an opportunity for enhanced transport during HVAC operation, and could be combined with other techniques, such as designing turbulent conditions near PRMs for further enhancement of mass transport. Some commercial HVAC technologies, such as ceiling mounted slot diffusers, have much higher velocities ($>1 \text{ m s}^{-1}$), and thus present additional opportunities for integration with PRMs.

Locating PRMs near ozone sources (both indoor sources and ozone transported from outdoors) and occupants are also strategies to improve PRM effectiveness. Ferro et al. (2009) found reductions of SF₆ between source and receptor rooms of 20%–99%, indicating significant spatial heterogeneity in pollutant concentrations in this residence. Because of this, PRMs should be preferentially placed in rooms with a higher probability of high ozone concentration, HVAC operation, and occupancy. Future research should incorporate time-activity patterns into PRM placement models to ensure that maximum pollutant removal occurs where occupants spend most of their time.

3.3. Surface area requirements

The extensive surface area available indoors represents both an opportunity and a challenge for the implementation of PRMs. As illustrated in Fig. 1, the GWB-High case results in higher effectiveness than ACC-Low. This is due to quadrupling the PRM surface area between GWB-High and ACC-Low compared to an increase of only 60% in deposition velocity between ACC-Low and GWB-High. Furthermore, at low and high air speeds, deposition velocity to ACC and GWB increases by only 13% and 2%, respectively, with an 80% increase in air speed. This is likely a result of the high air speeds associated with deposition velocities referenced in this study, a range where further increases in air speed result in small marginal increases in deposition velocity. Indoor comfort and energy expenditure are also concerns with high indoor air speeds, and illustrate the need to consider materials with potential for high surface area installations. This will allow design flexibility to optimize pollutant removal at lower indoor air speeds.

Incorporation of surface areas necessary to achieve minimum recommended effectiveness represents an important barrier to the implementation of PRMs. For widespread adoption in homes, PRMs must be made aesthetically pleasing to a wide demographic or engineered into common indoor materials. The lower surface area conditions presented (Low conditions), correspond to an average of 24 m^2 of PRM surface area in the Houston RIOPA homes. This is approximately $\frac{1}{3}$ – $\frac{1}{2}$ of the typical wall and ceiling area of a typical

bedroom. The High conditions presented in this work correspond to an average of 97 m^2 of PRM surface area in the Houston RIOPA homes. Future work should focus on researching materials that have known acceptability in indoor environments, as well as favorable pollutant scavenging properties. Inorganic materials such as perlite ceiling tiles and clay wall coverings are two promising PRMs commonly found in current building materials and that appear to be promising PRMs based on ongoing studies at the University of Texas. Novel PRM placement, such as incorporating PRMs into existing furnishings and surfaces or developing architectural coatings that allow reactions with underlying PRMs may be promising designs that could aid in achieving additional indoor surface area on the order of 10 – 100 m^2 .

3.4. Ozone-material reactions

PRMs present an opportunity for achieving a dual benefit of reduced indoor ozone concentrations and reduced formation of harmful ozone reaction byproducts. In the case of carpets, for example, Morrison and Nazaroff (2002) found secondary formation of aldehydes as high as $600 \mu\text{g m}^{-2} \text{ h}^{-1}$ for some carpets following exposure to 100 ppb of ozone. The formation of these aldehydes is hypothesized to result from the cleaving of carbon–carbon double bonds in oleic acid present as a result of the manufacturing process. Alternatively, in the case of activated carbon, the major ozone reaction products are oxygen, CO₂, H₂O, and small amounts of CO (Alvarez et al., 2008; Subrahmanyam et al., 2005). Optimizing removal of ozone to materials such as activated carbon will result in less ozone reacting with carpets and other materials, which emit harmful secondary products.

Other PRMs, such as GWB, pose a similar opportunity for reduced formation of ozone reaction byproducts, but illustrate the need for careful consideration of ozone reaction mechanisms. Nicolas et al. (2007) observed small increases in emission rates of aldehydes resulting from ozone reactions with GWB. Acetaldehyde emissions increased by $1 \mu\text{g m}^{-2} \text{ h}^{-1}$ following reaction with approximately 110 ppb of ozone. A conservative analysis in the median RIOPA home (volume = 271 m^3 , $\lambda = 0.5 \text{ h}^{-1}$), assuming a high GWB coverage ($0.3 \text{ m}^2 \text{ m}^{-3}$) and a very high indoor ozone concentration of 110 ppb (higher than reasonably anticipated), results in a predicted acetaldehyde emission rate of $97.2 \mu\text{g h}^{-1}$. This conservative upper bound would result in a steady-state indoor acetaldehyde concentration of $0.6 \mu\text{g m}^{-3}$ in the median RIOPA house, a value lower than the Environmental Protection Agency reference concentration (RfC) of $0.9 \mu\text{g m}^{-3}$ for this chemical (EPA, 2007). This RfC is a value that the EPA estimates as a safe level for a lifetime of continuous exposure. Regardless, this emission presents a trade-off: the benefits of reduced acute and chronic impacts associated with ozone exposure, as well as the prevention of other byproduct formation mechanisms that would occur as a result of reactions with other surfaces must be weighed against this increase in acetaldehyde concentration. In determining PRM viability, quantity and type of reaction byproducts must be well characterized prior to implementation.

3.5. Model limitations

There are a number of sources of uncertainty to consider in this analysis. Indoor emissions of ozone were not incorporated since all homes in areas of high outdoor ozone are affected, over which homeowners have limited control. Indoor sources of ozone exist, however are utilized in a small fraction of homes (California Air Resources Board, 2007) and typically have emission rates lower than the corresponding source strengths from outdoor ozone (Britigan et al., 2006). Furthermore, indoor ozone emissions can be

easily controlled by eliminating sources. Liu and Nazaroff (2001) reported a wide range of ozone penetration factors which would affect absolute exposure; however, no change to effectiveness will be realized as penetration factor affects the background and PRM cases similarly. Removal in the HVAC system was deemed of limited importance due to the removal capacity of HVAC filters of only 5%–25% for residential filters, likely a range of upper bounds due to low filter face velocity during these experiments (Zhao et al., 2007). Combined with typical residential HVAC operating times of 15 min per hour (Stephens et al., 2010), ozone removal on HVAC filters would not exceed 5%. This analysis also assumes that background removal is unaffected by the presence of PRMs, introducing a small bias if PRMs are assumed to cover low ozone deposition velocity surfaces, such as painted drywall (Grontoft and Raychaudhuri, 2004). Finally, material reactivity with ozone may change with respect to time. Reductions in material reactivity would reduce the calculated values of effectiveness below those presented in this investigation.

4. Conclusions

The known health effects associated with ozone exposure coupled with the documented temporal importance of the indoor environment warrant investigation into low-energy methods for indoor ozone control. The use of passive indoor controls will allow building designers to improve the quality of indoor air by capitalizing on existing building phenomena to reduce or eliminate energy inputs typically required for air purification. Passive removal materials will also allow for ozone control to be readily deployed to susceptible populations where the health benefits are greatest. To further the aim of creating healthy indoor environments, the exploration of PRMs that remove or sequester other common indoor pollutants such as particles, microbiological contaminants, and VOCs is warranted.

Acknowledgment

Elliott Gall was supported by a National Science Foundation IGERT traineeship (Award 0549428).

References

- Alvarez, P.M., Masa, F.J., Jaramilla, J., Beltrn, F., Gomez-Serrano, V., 2008. Kinetics of ozone decomposition by granular activated carbon. *Industrial & Engineering Chemistry Research* 47 (8), 2545–2553.
- Bell, M., Dominici, F., Samet, J., 2005. A meta-analysis of time-series studies of ozone and mortality with comparison to the national morbidity, mortality, and air pollution study. *Epidemiology* 16 (4), 436–445.
- Britigan, N., Alshawa, A., Nizkorodov, S.A., 2006. Quantification of ozone levels in indoor environments generated by ionization and ozonolysis air purifiers. *Journal of the Air and Waste Management Association* 56, 601–610.
- California Air Resources Board, 2007. Survey of the use of ozone generating air cleaners by the California public. CARB, Sacramento.
- DOE, 2006. Buildings Energy Data Book. 2006 Building Energy End-Use Splits. Retrieved October 1, 2010. <http://buildingsdatabook.eren.doe.gov/ChartView.aspx?chartID=0>.
- EPA, 2007. Technology Transfer Network Air Toxics Web Site. Retrieved April 5, 2010. <http://www.epa.gov/ttn/atw/hlthef/acetalde.html>.
- Ferro, A., Klepeis, N.E., Ott, W.R., Nazaroff, W.W., Hildemann, L.M., Switzer, P., 2009. Effect of interior door position on room-to-room differences in residential pollutant concentrations after short-term releases. *Atmospheric Environment* 43, 706–714.
- Grontoft, T., Raychaudhuri, M., 2004. Compilation of tables of surface deposition velocities of O₃, NO₂, and SO₂ to a range of indoor surfaces. *Atmospheric Environment* 38, 533–544.
- Kunkel, D.A., Gall, E.T., Siegel, J.A., Novoselac, A., Morrison, G.C., Corsi, R.L., 2010. Passive reduction of human exposure to indoor ozone. *Building and Environment* 45, 445–452.
- Lee, K., Vallarino, J., Dumyahn, T., Ozkaynak, H., Spengler, J.D., 1999. Ozone decay rates in residences. *Journal of the Air and Waste Management Association* 49 (10), 1238–1244.
- Liddament, M., Orme, M., 1998. Energy and ventilation. *Applied Thermal Engineering* 181 (11), 1101–1109.
- Liu, D.-L., Nazaroff, W.W., 2001. Modeling pollutant penetration across building envelopes. *Atmospheric Environment* 35, 4451–4462.
- Matthews, T.G., Thompson, C.V., Wilson, D.L., Hawthorne, A.R., Mage, D.T., 1989. Air velocities inside domestic environments: an important parameter in the study of indoor air quality and climate. *Environment International* 15, 545–550.
- Miller-Leiden, S., Lobascio, C., Nazaroff, W.W., Macher, J.M., 1996. Effectiveness of in-room air filtration and dilution ventilation for tuberculosis infection control. *Journal of the Air and Waste Management Association* 46 (9), 869–882.
- Morrison, G.C., Nazaroff, W.W., 2002. Ozone interactions with carpet: secondary emissions of aldehydes. *Environmental Science & Technology* 36, 2185–2192.
- Nicolas, M., Ramalhoa, O., Maupeti, F., 2007. Reactions between ozone and building products: impact on primary and secondary emissions. *Atmospheric Environment* 41 (15), 3129–3138.
- Sekine, Y., Nishimura, A., 2001. Removal of formaldehyde from indoor air by passive type air-cleaning materials. *Atmospheric Environment* 35, 2001–2007.
- Shaughnessy, R.J., Sextro, R.G., 2006. What is an effective portable air cleaning device? A review. *Journal of Occupational and Environmental Hygiene* 3, 169–181.
- Stephens, B., Siegel, J.A., Novoselac, A., 2010. Energy implications of filtration in residential and light-commercial buildings (RP-1299). *ASHRAE Transactions* 116 (1), 346–357.
- Subrahmanyam, C., Bulushev, D.A., Kiwi-Minsker, K., 2005. Dynamic behavior of activated carbon catalysts during ozone decomposition at room temperature. *Applied Catalysis B: Environmental* 61, 98–106.
- Weisel, P., Zhang, J., Turpin, B., Morandi, M., Colome, S., Stock, T., Spektor, D., Korn, L., Winer, A., Kwon, J., Meng, Q., Zhang, L., Harrington, R., Liu, W., Reff, A., Lee, J., Alimokhtari, S., Mohan, K., Shendell, D., Jones, J., Farrar, L., Maberti, S., Fan, T., 2005. Research Report - Health Effects Institute, 1–107.
- Weschler, C.J., 2000. Ozone in indoor environments: concentration and chemistry. *Indoor Air* 10, 269–288.
- Weschler, C.J., 2006. Ozone's impact on public health: contributions from indoor exposures to ozone and products of ozone-initiated chemistry. *Environmental Health Perspectives* 114 (10), 1489–1496.
- Yamamoto, N., Shendell, D.G., Winer, A.M., Zhang, J., 2010. Residential air exchange rates in three major US metropolitan areas: results from the relationship among indoor, outdoor and personal air study 1999–2001. *Indoor Air* 20, 85–90.
- Zhao, P., Siegel, J.A., Corsi, R.L., 2007. Ozone removal by HVAC filters. *Atmospheric Environment* 41, 3151–3160.