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Controlling Indoor Bioaerosols Using a Hybrid System of Ozone and Catalysts

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ABSTRACT

As people spend a greater portion of their time indoors, the likelihood of exposure to biohazards in indoor air is increasing. This study developed a system to disinfect indoor bioaerosols while protecting occupants from direct exposure to ozone. This hybrid approach combines high concentrations of ozone generated by a non-thermal dielectric barrier discharge system, and an ozone decomposition unit using MnO_2/Al_2O_3 and MnO_2/AC catalysts. Four types of common bioaerosols were used to test the germicidal efficacy of 0–175 ppm ozone at a relative humidity of 30–70% and retention time of 1–10 s. The results indicate that the germicidal efficacy of ozone on *E. coli, C. famata* and *P. citrinum* spore bioaerosols increased with ozone concentration, relative humidity, and retention time; however, the process was less effective with the hardy endospores of *B. subtilis*. Germicidal efficacy of 90% was obtained for bioaerosols (two vegetative cells and one spore) using high concentrations of ozone at a relative humidity of 70% and exposure time of 10 s. Residual ozone in the tail gas was decomposed to undetectable levels using catalysts at a gas hourly space velocity of 3.287 × 10³ 1/h. The concentration of indoor bioaerosols (total bacteria and fungi) in a small office was reduced to below 50 CFU/m³ and ozone in the tail gas was reduced to undetectable levels following treatment for 2 h. The hybrid system of ozone disinfection and catalysts provides high germicidal efficacy while protecting occupants from direct exposure to ozone.

Keywords: Disinfection; Indoor air quality; Biohazard; Germicidal efficacy; Microorganism.

INTRODUCTION

Many diseases, such as tuberculosis, severe acute respiratory syndrome (SARS), and H1N1 flu are caused by airborne microbes. Modern lifestyles involve people spending more than 80% of their time indoors (Sundell, 2004), which increases the chance of exposure to biohazards in indoor environments. The adverse effects of bioaerosols include infectious diseases, acute toxic effects, allergies, and cancer. Among these, infectious respiratory diseases are the most prevalent (Douwes *et al.*, 2003; Ezzell *et al.*, 2009; Lee *et al.*, 2009).

The subtropical climate of Taiwan, with high temperature and humidity year round, is well suited to the growth of microorganisms. Many studies have identified high levels of bacteria, fungi, and allergens in indoor environments in Taiwan. Li and Kuo (1992; 1993) indicated that temperature and relative humidity are related to the concentration of

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fungal bioaerosols in homes in northern Taiwan. Li *et al.* (1997) investigated the most common house dust mite Der P 1 allergens present in mattresses during the summer and winter. Su *et al.* (2001a) revealed that the levels of fungal bioaerosols in homes and schools in southern Taiwan appear higher than those in northern Taiwan and other countries (Su *et al.*, 2001b). Wu *et al.* (2005) found that higher indoor/ outdoor ratios of fungi and bacteria exist in buildings with fan coil units compared to those with air-handling units. Li *et al.* (1996) demonstrated the prevalence of dampness in daycare centers Taiwan, resulting in concentrations of indoor bacteria and fungi exceeding outdoor levels.

Many aerobiological technologies have been developed to control indoor bioaerosols: ventilation, filtration, ultraviolet germicidal irradiation, photocatalytic oxidation, ionization, ozone, negative ion, thermal disinfection (Christopher and Pasquale, 2002; Whitney *et al.*, 2003; Kowalski, 2006; Tseng and Li, 2006; Yu *et al.*, 2008; Yoosook *et al.*, 2009). Ozone is a strong oxidant with a tremendous ability to oxidize odors, organic compounds, microorganisms, and other substances (Franken, 2005). Due to its powerful oxidation potential, ozone has been applied in the disinfection of microorganisms in many fields, such as food, water, air, soil, and medicine (Franken, 2005; Takayama *et al.* 2006; Tseng and Li,

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2006; Naitou and Takahara, 2008; Uhm *et al.*, 2009). Gaseous ozone is particularly effective in odor removal, is largely non-corrosive to equipment, has excellent disinfection capability, and provides complete coverage of all surfaces, making it ideal for application as an interior disinfectant (Franken, 2005).

Ishizaki et al. (1986) indicated that following surface ozone disinfection, the inactivation rates of six strains of Bacillus spores on filter paper or glass fiber filter increased with relative humidity. Li and Wang (2003) showed that ozone had high surface disinfection efficiency for a wide range of bacteria and fungi, except for Bacillus subtilis. Kowalski et al. (2003) used low concentrations of ozone to disinfect Escherichia coli. on petri dishes demonstrating death rates of microorganisms exceeding 99.99%. Klánová and Lajcíková (2005) also found that ozone was effective in reducing Escherichia coli and Pseudomonas aeruginosa on slides. Ozone outperforms UV radiation for disinfecting microorganisms. Akbas and Ozdemir (2008) revealed that vegetative cells could be reduced at 1 ppm of ozone in dried figs; however, Bacillus cereus spores required higher concentrations. Najafi and Khodaparast (2009) showed that Escherichia coli and Staphylococcus aureus could not be cultured in date fruits following treatment with 5 ppm ozone for 60 min.

Serra *et al.* (2003) used ozone to reduce molds to below the level of 50 MPN/m³ in a cheese ripening room over a 3-month period. Ozone has proven to be an effective method to inactivate molds in a closed space. Wang (2003) found that low concentrations of ozone have high germicidal efficacy against *Escherichia coli* bioaerosols, but low germicidal efficacy for *yeast* bioaerosols in chamber studies. Klánová and Lajcíková (2005) indicated that the levels of *Escherichia coli* and *Pseudomonas aeruginosa* bioaerosols decreased to undetectable levels following ozone treatment for 12 h. Tseng (2006) found that ozone is highly effective in disinfecting four airborne viruses; however, viruses are generally difficult to treat with ozone because of their more complex capsid architectures and short contact time.

Because ozone is harmful to humans, few studies have applied gaseous ozone to disinfect bioaerosols. Ozone is a powerful oxidant capable of reacting with organisms and damaging the respiratory system even at low concentrations. Ozone can cause chest pain, coughing, shortness of breath, throat irritation, and impairment of lung function (Delfino et al., 1996; Franken, 2005; US EPA, 2011). Exposure to ozone is associated with mortality, morbidity, respiratory symptoms, and loss of productivity (Weschler, 2006). To prevent direct exposure to ozone, the application of this technology is limited to confined, unoccupied spaces. Unfortunately, this greatly limits the applicability of gaseous ozone for disinfection in indoor environments. Many studies (Dhandapani and Oyama, 1997; Naydenov et al., 2008; Yu et al., 2009) have indicated that catalysts comprising noble metals or oxides of metals and catalyst supports (e.g. γ - Al_2O_3 , SiO_2 , TiO_2 and activated carbon) are able to decompose ozone with a high degree of efficiency.

Another approach to reducing the harmful effects of ozone is to reduce the concentration and treatment time for bioaerosol disinfection. However, this lowers disinfection efficiency. The purpose of this study was to develop a new continuous ozone disinfection system comprising high concentrations of ozone generated by non-thermal plasma with catalysts for the decomposition of ozone. This combined approach enables the use of ozone for indoor disinfection, while avoiding direct exposure.

This study estimated the germicidal effectiveness of the disinfection system on bacterial and fungal bioaerosols at various ozone concentrations, degrees of relative humidity, and treatment times. In addition, a field study was performed to assess the feasibility of using the ozone disinfection system to reduce bioaerosols and improve indoor air quality in a small office.

METHODS

Ozone Generation Unit

High concentrations of ozone were generated using a dielectric barrier discharge (DBD) plasma reactor. The DBD plasma reactor consisted of a dielectric barrier and inner and outer electrodes. The dielectric barrier was a glass tube 35 cm long and 3.6 cm in diameter. The inner electrode was a rod of stainless steel 0.32 cm in diameter. The outer electrode was a wire mesh 29 cm long. One of two electrodes in the DBD plasma reactor was grounded and the other was connected to a high voltage transformer, which was controlled by a plasma controller (Model: CF-500T, Chung Lin Electric Industrial Co., Taiwan), to adjust the voltage, frequency, and power. The high voltage generated by the transformer was transferred to the inner electrode. Ozone was generated using a specified input voltage.

Bioaerosol Species

Spore-type microorganisms are typically hardier than their non-spore counterparts in indoor environments. Two bacteria and fungi species were selected as test microorganisms to assess the germicidal effects of ozone on bacterial and fungal bioaerosols and compare the differences in the effectiveness of treatment between species. The bacterial bioaerosols were vegetative cells of Escherichia coli (E. coli, CCRC 10675, Culture Collection & Research Center in Taiwan) and endospores of Bacillus subtilis (B. subtilis, CCRC 12145) and the test fungal bioaerosols were vegetative cells of Candida famata (C. famata, CCRC 22304), and spores of Penicillum citrinum (P. citrinum, Thom CCRC 33168). The genus, C. famata, is a common airborne yeast isolate in Taiwan, identified by the Taiwan Food Industrial Research and Development Institute. P. citrinum is also a common genus isolate in Taiwan.

Catalytic Decomposition Unit of Ozone

Two commercial catalysts, MnO_2/Al_2O_3 and $MnO_2/Activated$ Carbon (AC), were purchased from Lanzhou Institute of Chemical Physics (Chinese Academy of Science, China) to decompose ozone in this study. The sizes of the rod shaped catalysts were approximately 2–3 mm (diameter) \times 5–10 mm (length). The densities of the two catalysts were approximately 0.45–0.5 g/cm³. The average life of

these catalysts was estimated at approximately 8000 h. The suitable operation temperature was $20-30^{\circ}$ C, the ozone decomposition ability was lower than 0.1 ppm of ozone concentration after decomposition, and the optimal height to diameter ratio of the catalyst bed was 4-8:1. The catalysts were packed into a tube of dimensions 4.4 cm (diameter) × 24 cm (length). The tube containing the catalysts was connected to the outlet of the ozone disinfection chamber to treat residual ozone in the tail gas following disinfection. The efficacy of the catalytic decomposition of ozone was calculated by measuring the ozone concentration in the ozone disinfection chamber and the outlet of the catalyst bed.

Ozone Disinfection in Chamber

Fig. 1 presents a schematic of the ozone disinfection system for bioaerosols. The bioaerosols were atomized from a microorganism suspension using a Collision Threejet Nebulizer (BGI, Inc., USA) and passed through a diffusion dryer (Model 306200, TSI Inc., USA) to remove excess moisture. The bioaerosols were then passed through a Kr-85 radioactive source (model 3077, TSI Inc., USA), which neutralized the aerosol to the Boltzmann charge equilibrium. Test bioaerosols were mixed with gaseous ozone, humid air, and diluted clean air to a specific relative humidity (RH) prior to injection into the ozone disinfection chamber. Finally, the disinfected airflow passed through a catalytic decomposition unit before being discharged.

The concentration of the test bioaerosols was approximately 10^6 CFU/m³. The concentration of ozone for disinfection was 0–175 ppm. The relative humidity was controlled at 30, 50, and 70%. The retention times of ozone disinfection were 1.5, 5, and 10 s. The levels of ozone in the disinfection chamber and outlet of the catalytic

decomposition unit were measured using a Model 205 Dual Beam ozone monitor (2B Technologies, USA). An Anderson single-stage bioaerosol impactor (SKC Product Code: 225-9611, USA) equipped with Tryptic Soy Agar (TSA) and Malt Extract Agar (MEA) plates was used to collect bacteria and fungi prior to and following the disinfection process, by turning the ozone generation unit off and on. Triplicate samples of bacterial and fungal bioaerosols were collected at a flow rate of 28.3 L/min for 5-10 s for each set of experimental conditions, and all disinfection experiments were repeated three times. After sampling, TSA and MEA plates were cultivated at 30°C and 25°C for 2 days and 5 days, respectively. The numbers of colony forming units of bacteria and fungi in each plate were counted to calculate bioaerosol concentrations (CFU/m³). Finally, the germicidal efficacy of ozone was calculated according to the bioaerosol concentrations before and after disinfection.

Ozone Disinfection in Field Study

The ozone disinfection system was set up in a small office with a split air conditioner at a temperature of approximately 25°C and a relative humidity of 50–55%. The sites of the ozone disinfection system and bioaerosol sampling are shown in Fig. 2. The size of the office was 4.2 m (length) \times 3.0 m (width) \times 2.9 m (height). The units of the ozone disinfection system were the same as those in Fig. 1, except that the source of bioaerosols was the air of the office. Indoor air was continuously delivered into the ozone disinfection chamber for contact with the ozone using an air pump. The retention time was 10 s, the ozone concentration was 150 ppm, and the total gas flow rate was 20 L/min. The disinfected air was transferred into the MnO₂ catalyst unit to decompose ozone prior to discharge into the indoor atmosphere. The duration of continuous disinfection



Fig. 1. Ozone disinfection system for bioaerosols.



Fig. 2. Layout of ozone disinfection system in office.

was 2 h. Bacterial and fungal bioaerosols were collected every 10 min. Ozone levels at the outlet of the catalytic decomposition unit were measured continuously using an ozone monitor. Following cultivation, the numbers of colony forming units of bacteria and fungi in each plate were counted to calculate the total concentrations of airborne bacteria and fungi (CFU/m³). Due to on-going variations in the concentrations of indoor bioaerosol, individual experiments under the two ventilation conditions of air-conditioning off and on were conducted on different days.

RESULTS AND DISCUSSION

Effects of Ozone Concentrations

The germicidal efficacy of ozone on four different bioaerosols at an RH of 50% and retention time of 10 s are shown in Fig. 3. The germicidal efficacy of ozone on E. coli, C. famata, and P. citrinum spores increased with ozone concentration. The germicidal efficacy of ozone on E. coli, C. famata, and P. citrinum spore bioaerosols reached approximately 95%, 81%, and 71% at maximum ozone concentrations of 50 ppm, 100 ppm, and 150 ppm, respectively. Wang (2001) indicated that a low ozone concentration of 6 ppm had high germicidal efficacy for E. coli bioaerosols at an RH of 55% and exposure time of 18.4 s. Although the experimental conditions of Wang (2001) differed from those in this study, this study confirms the high germicidal efficacy of gaseous ozone on E. coli bioaerosols. However, the germicidal efficacy of ozone on C. famata bioaerosols in both studies were lower than 10% at low ozone concentrations of 10 ppm and 20 ppm (RH = 55% and exposure time = 13.8 sec). Unlike E. coli bioaerosols with high germicidal efficacies at low ozone concentrations,

the germicidal efficacy of ozone on *C. famata* bioaerosols increased markedly only at concentrations above 100 ppm.

Fig. 3 shows that the germicidal efficacy of ozone on P. citrinum bioaerosols was lower than 30% at concentrations below 100 ppm. Li and Wang (2003) discovered that surface disinfection of hardy P. citrinum spores required extended exposure to ozone for 2 h at a concentration of 12 ppm to reduce the survival fraction to below 0.4. This implies that high ozone concentrations are required to disinfect P. citrinum spores. However, Fig. 3 shows that gaseous ozone has no germicidal effect on the endospore bioaerosols of B. subtilis even at a high ozone concentration of 175 ppm and retention time of 10 s. Endospores of B. subtilis have a very high tolerance to ozone. These results are similar to those of Ishizaki et al. (1986), who used high ozone concentrations of 0.5-3.0 mg/l to disinfect endospores of B. subtilis on filter paper. The germicidal efficacy of ozone on B. subtilis bioaerosols at a high ozone concentration of 175 ppm was not observed in this study, due possibly to the endospores of *B. subtilis* remaining within the lag phase, which showed no change in the number of survivors during the short retention time of 10 s (Ishizaki et al., 1986). This suggests that higher ozone concentrations and prolonged disinfection times are probably required for inactive airborne endospores of B. subtilis.

The results in Fig. 3 clearly illustrate the trend of germicidal efficacy using high ozone concentration for four bioaerosols: *E. coli* > *C. famata* > *P. citrinum* spores > endospores of *B. subtilis* (i.e., the order of tolerance to ozone: endospores of *B. subtilis* > *P. citrinum* spores > *C. famata* > *E. coli*). The tolerance of (cell-type) fungal *C. famata* to high concentrations of ozone was better than bacterial *E. coli*. These results are consistent with those of



Fig. 3. The germicidal efficacy of ozone on bioaerosols at an RH of 50% and retention time of 10 s.

Li and Wang (2003) with regard to surface disinfection using low concentrations of ozone. The lipopolysaccharide layer in the cell membrane of gram-negative E. coli was the main reaction site of the ozone. However, the general thickness of the cell walls of fungi is approximately 100-200 nm. The cell wall could act as a barrier layer and delay the penetration of ozone into the cell membrane (Moore et al., 2000). Without the protection of the barrier, E. coli bioaerosols were more susceptible to ozone than C. famata bioaerosols. However, the tolerance of bacterial endospores of B. subtilis to high concentrations of ozone was better than fungal P. citrinum spores. Endospores of B. subtilis were obviously hardier than P. citrinum spores at high concentrations of ozone. These results are similar to those of Wang (2001) using low ozone concentrations to disinfect bioaerosols and the ozone disinfection of surfaces by Li and Wang (2003). For both bacteria and fungi, the tolerance of spore-type bioaerosols to high concentrations of ozone was better than cell-type bioaerosols. B. subtilis and P. citrinum, which required higher concentrations of ozone than E. coli or C. famata to be sterilized. This was due to the protection provided by the multi-shell structure of spores. These results are consistent with those of Wang (2001), Li and Wang (2003) and Kammer (2005).

Effects of Relative Humidity

The germicidal efficacy of ozone on bioaerosols, at various RHs with a retention time of 10 s, is shown in Fig. 4. The results indicate that the germicidal efficacy of ozone on *E. coli*, *C. famata*, and *P. citrinum* spores increased with RH. Because *E. coli* has a low tolerance to ozone, the germicidal efficacy of *E. coli* bioaerosols reached 95% at an RH of 50%. The germicidal efficacy of *E. coli* bioaerosols increased approximately 17% when the RH increased from 30% to 50%. Therefore, the increase in germicidal efficacy

was relatively limited when the RH was further increased from 50% to 70%. For *C. famata* bioaerosols, germicidal efficacy increased nearly linearly from 65% at an RH of 30% to reach 99% at an RH of 70%. The increase in the germicidal efficacy of ozone on *C. famata* bioaerosols by increasing the RH was significant. The germicidal efficacy of ozone on *P. citrinum* spore aerosols was only 13% at an RH of 30%; however, it increased to 71% and 95% when the RH was increased to 50% and 70%, respectively. This demonstrated that a high RH was very helpful in disinfecting *P. citrinum* spore bioaerosols. Similar to previous results on the effects of ozone concentration, RH did not influence the disinfection of airborne endospores of *B. subtilis*.

One possible reason for the increase in the germicidal efficacy of ozone on the three bioaerosols of E. coli, C. famata, and P. citrinum spores was the quantity of hydroxyl radicals generated by the reaction of ozone with H₂O at an elevated RH (Li and Wang, 2003; Kowalski, 2006). The generation of hydroxyl radicals of high activity further interacted with bioaerosls to inactive the cells of microorganisms. However, the increase in germicidal efficacy on C. famata bioaerosols was not observed by Wang (2001). This could be due to the 10 ppm ozone concentration used, which was too low to generate sufficient hydroxyl radicals to inactive C. famata. Ozone concentrations needed to be elevated to observe the effect of RH on disinfection. Regarding the endospores of B. subtilis, no germicidal effects presented even at a high RH of 70% with an ozone concentration of 175 ppm. Hydroxy radicals were unable to enhance the efficiency of ozone disinfection for hardy B. subtilis bioaerosols. Although the formation of hydroxyl radicals did not enhance the effectiveness in this study, Ishizak et al. (1986) found that an increased RH could decrease the survival rate of endospores of B. subtilis on filter paper following extended disinfection with



Fig. 4. The germicidal efficacy of ozone on bioaerosols at various relative humidities (retention time = 10 sec; ozone concentration-*E. coli*: 50 ppm; endospores of *B. subtilis*: 175 ppm; *C. famata*: 100 ppm; *P. citrinum* spores: 150 ppm).

high concentrations of ozone. It was speculated that RH may play an important role in facilitating the germicidal effect only when gaseous ozone is used for the disinfection of microorganisms, such as *E. coli*, *C. famata*, and *P. citrinum* at ozone concentrations of 50ppm, 100ppm, and 150ppm, respectively. Otherwise, the results reveal that the germicidal efficacy of high concentrations of ozone on bioaerosols exceed 95% at an RH of 70%, except for *B. subtilis*, as shown in Fig. 4. These results suggest that high concentrations of ozone would be very suitable for the disinfection of airborne microorganisms in Taiwan, in which the environment has a high RH year round.

Effects of Retention time

The germicidal efficacy of ozone on bioaerosols varied with retention time in the ozone disinfection chamber, as shown in Fig. 5. These results indicate that the germicidal efficacy of ozone on E. coli, C. famata, and P. citrinum spore bioaerosols increased with retention time, except for the endospores of B. subtilis. Gaseous ozone demonstrated no disinfection capability on the endospores of *B. subtilis*, even when the retention time was extended to 10 s. The longer retention time, the higher was the germicidal efficacy. It has been suggested that prolonged retention time could increase the probability of exposure between ozone and bioaerosols and provide sufficient time for the ozone to diffuse into the microorganism to oxidize the cells. The germicidal efficacy of ozone on E. coli, C. famata, and P. citrinum spore bioaerosols increased noticeably when retention time was increased from 1.5 s to 5.0 s. Similar results were obtained in ozone surface disinfection by Wang (2001) and Li and Wang (2003). However, only a slight increase in germicidal efficacy was observed with a further increase in retention time from 5.0 s to 10 s. The

germicidal efficacy of ozone on *E. coli* bioaerosols exceeded 90% at a retention time of 5.0 s; therefore, the enhancement of germicidal efficacy following an increase in retention time to 10 s was limited.

The germicidal efficacy of ozone on C. famata and P. citrinum spore bioaerosols was approximately 80% and 70% with a retention time of 5.0 s. Changes in germicidal efficacy following an increase in retention time to 10 s were also limited, demonstrating results similar to those observed with E. coli bioaerosols. According to previous results, germicidal efficacy exceeded 95% when the RH was increased to 70%. This implies that such as short increase in retention time would be unable to reveal the influence of disinfection time on germicidal efficacy. On the contrary, the contribution of RH to germicidal efficacy exceeded that of retention time. Relative to retention time, RH was definitely a more important factor in enhancing the germicidal efficacy of ozone on bioaerosols, largely because hydroxyl radicals are generated at a high RH. Ishizaki et al. (1986) demonstrated that the spores of B. subtilis on filter paper at an RH of 50% required many hours to achieve inactivation, even when using high concentrations of ozone. Furthermore, the bioaerosol disinfection of the endospores of B. subtilis using ozone differed very little from the ozone disinfection of surfaces. The probability of exposure between bioaerosols and gaseous ozone was lower than that of microorganisms on surfaces and ozone. Therefore, even with a high concentration of ozone, disinfection of Bsubtilis bioaerosols undoubtedly still required time for the ozone and radicals to infiltrate the hardy multi-shells of the endospores. Observing the germicidal efficacy of ozone on endospore bioaerosols of B. subtilis with a short retention. time of only 10 s (the duration established for this study) was not possible.



Fig. 5. The germicidal efficacy of ozone on bioaerosols at various retention times (RH = 50%; ozone concentration-*E. coli*: 50 ppm; endospores of *B. subtilis*: 175 ppm; *C. famata*: 100 ppm; *P. citrinum* spores: 150 ppm).

Efficacy of Catalytic Decomposition of Ozone

To ensure that the residual gaseous ozone discharged from the ozone disinfection chamber was not harmful to residents, two catalysts, MnO₂/Al₂O₃ and MnO₂/AC, were used to treat the ozone in the tail gas. The ratio of height to diameter of the catalyst bed was 5.5:1. Test results show that ozone used for disinfecting bioaerosols (50-175 ppm) was decomposed to undetectable levels by the catalysts at a gas hourly space velocity (GHSV) of 1.03×10^4 1/h. The precision of ozone monitoring used in this study was 0.01 ppm. It was estimated that the residual ozone in the tail gas decomposed to less than 10 ppb when using either of the two catalysts. The concentration of undetectable ozone discharged from the catalyst bed was lower than the indoor air quality (IAQ) limit recommended by the Taiwan EPA (O3: 0.03 ppm-8 h average) (Taiwan EPA, 2005) and other standards from the USA (FDA: 0.05 ppm; OSHA: 0.10 ppm-8 h average; NIOSH: 0.10 ppm-any time; NAAQ: 0.08 ppm-maximum 8 h average) (US EPA, 1999), which suggests that the potential adverse health effects of exposure to such levels of ozone are insignificant. The results of efficacy of the two catalysts in the decomposition of ozone are similar to those of Dhandapani and Oyama (1997) and Yu et al. (2009). It was demonstrated that catalytic decomposition is suitable for treating high concentrations of residual ozone discharged from the ozone disinfection system if the catalysts are selected correctly.

Ozone disinfection in small office

Previous results have indicated that 150 ppm ozone at an RH of 50% and retention time of 10 s had a germicidal efficacy of at least 70% on *E. coli*, *C. famata*, and of *P. citrinum spore* bioaerosols. Therefore, the operational conditions of the ozone disinfection system in the field

study were set up at an ozone concentration of 150 ppm, retention time of 10 s, and air flow rate of 20 L/min. The air change per hour (ACH) in the office was determined by emitting high concentrations of CO₂ into the office and detecting the variation of CO₂ concentration using a Q-Trak CO₂ monitor (model 8554, TSI Inc., USA). The ACH of the office with air-conditioning off and on were 0.45 1/h and 3.89 1/h, respectively. Fig. 6 shows the variations in total indoor bacterial and fungal bioaerosol concentrations in the office. The results indicate that bioaerosol concentrations gradually decreased with an increase in germicidal time. Regardless of whether the split air conditioner was turned on or off in the office, the levels of total indoor bacterial and fungal bioaerosols decreased below 50 CFU/m³ following 2 h of continuous disinfection. The numbers of airborne bacteria and fungi were both abated effectively under the treatment of ozone disinfection system.

Wang (2001) emitted low concentrations of ozone (1 ppm and 10 ppm) directly into a room to explore the germicidal efficacy on indoor bioaerosols over a period of 1.5 h. The results show that 1 ppm ozone had no effect on reducing bioaerosols, regardless of whether the ventilation system was turned on or off. The total decrease in bacterial and fungal bioaerosols using 10 ppm of ozone was approximately only 50 CFU/m³ at a distance of 1 m from the ozone source. No germicidal efficacy was observed for bioaerosols after turning on the ventilation system (ACH = 3.5 1/h). These results clearly indicate that air disinfection by ozone is influenced by ozone concentration, distance, and ventilation. However, the ozone disinfection system in this study was similar to an air cleaner. Indoor air was continuously pumped into a disinfection chamber and treated with 150 ppm of ozone for a period of 10 s. The residual ozone in effluent air was then completely decomposed using catalysts and



Fig. 6. The variations of bioaerosol concentrations in office with (a) air-conditioning off (ACH = 0.45 l/h) and (b) air-conditioning on (ACH = 3.89 l/h).

discharged back into the indoor air. Thus, the controlled concentration of ozone and retention time in this continuous ozone disinfection system were constant and unaffected by the ventilation system.

The results of this field study show that the germicidal efficacy of the ozone disinfection system for indoor bacterial and fungal bioaerosols was above 80% and 70%, respectively, after continuous disinfection for 2 h. The germicidal efficacy of ozone measured at an ACH of 0.45 1/h and 3.89 1/h, indicating that the effect of ventilation on ozone disinfection system was negligible. Owing to limits on the treatment capacity, this small ozone disinfection system was used only in a small office. Nonetheless, the results demonstrate that the ozone disinfection unit connected with catalysts has good bioaerosol germicidal efficacy and high efficiency

in the decomposition of ozone to protect occupants in the office from exposure to ozone.

CONCLUSIONS

The study developed an ozone disinfection system to explore the germicidal efficacy of ozone on indoor bioaerosols. The results demonstrate that high concentrations of gaseous ozone have good germicidal efficacy for three bioaerosol species, *E. coli*, *C. famata*, and spores of *P. citrinum*; however, it was less effective against the hardy endospores of *B. subtilis*. An increase in the concentration of ozone, relative humidity, or retention time was helpful to enhance the germicidal efficacy of ozone on the three species of bioaerosols. The germicidal efficacy of ozone on cell-type bioaerosols was higher than that of sporeforming bioaerosols. Residual ozone in the tail gas was decomposed to undetectable levels through the use of catalysts. Testing the ozone disinfection system in a small office demonstrated good germicidal efficacies for indoor bioaerosols with no ozone detected at the outlet of the catalytic decomposition unit under various ventilation conditions. In the past, low concentrations of ozone over extended disinfection periods were required to obtain good germicidal efficacy. Under such conditions, occupants were also required to evacuate the premises to avoid direct exposure to harmful ozone. The results in this study demonstrate that an ozone disinfection system, comprising an ozone disinfection chamber and catalytic decomposition unit using high concentrations of ozone with a short retention time, is an effective disinfection method for the control of indoor bioaerosols. This system also protects occupants from exposure to ozone. Preliminary results in this study reveal that this ozone disinfection system has considerable potential for the control of indoor airborne bioaerosols.

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REFERENCES

- Akbas, M.Y. and Ozdemir, M. (2008). Application of Gaseous Ozone to Control Populations of Escherichia Coli, Bacillus Cereus and Bacillus Cereus Spores in Dried Figs. *Food Microbiol.* 25: 386–391.
- Brosseau, L.M., Evans, J.L., Ellenbecker, M.J. and Feldstein, M.L. (1989). Collection Efficiency of Respirator Filters Challenged with Monodisperse Latex Aerosols. *Am. Ind. Hyg. Assoc. J.* 50: 544–549.
- Christopher, F.G. and Pasquale, V.S. (2002). The Use of Ultraviolet Germicidal Irradiation (UVGI) in Disinfection of Airborne Bacteria. *Environ. Eng. Policy* 3: 101–107.
- Delfino, R.J., Coate, B.D., Zeiger, R.S., Seltzer, J.M., Street, D.H. and Koutrakis, P. (1996). Daily Asthma Severity in Relation to Personal Ozone Exposure and Outdoor Fungal Spores. Am. J. Respir. Crit. Care Med. 154: 633–641.
- Dhandapani, B. and Oyama, S.T. (1997). Gas Phase Ozone Decomposition Catalysts. *Appl. Catal., B.* 11: 129–166.
- Douwes, J., Thorne, P., Pearce, N. and Heederik, D. (2003). Bioaerosol Health Effects and Exposure Assessment: Progress and Prospects. *Ann. Occup. Hyg.* 47: 187–200.
- Ezzell, J.W., Abshire, T.G., Panchal, R., Chabot, D., Bavari, S., Leffel, E.K., Purcell, B., Friedlander, A.M. and Ribot, W.J. (2009). Association of Bacillus Anthracis Capsule with Lethal Toxin during Experimental Infection. *Infect. Immun.* 77: 749–755.
- Franken, L. (2005). The Application of Ozone Technology for Public Health and Industry, Food Safety & Security at Kansas State University, p. 1–16.
- Ishizakin, K., Inrik, A. and Matsuyama, H. (1986). Inactivation

of Bacillus Spores by Gaseous Ozone. J. Appl. Bacteriol. 16: 67–72.

- Klánová, K. and Lajcíková, A. (2006). Use of Ozone to Reduce Bacteria and Moulds in the Air and on Surfaces. *Indoor Built Environ.* 15: 81–84.
- Kowalski, W.J. (2006). Aerobiological Engineering Handbook: A Guide to Airborne Disease Control Technologies, New York, McGraw-Hill.
- Kowalski, W.J., Bahnfleth, W.P., Striebig, B.A. and Whittamc, T.S. (2003). Demonstration of a Hermetic Airborne Ozone Disinfection System: Studies on *E. Coil. AIHA J.* 64: 222–227.
- Lee, J.E., Fang, X., Gupta, N., Serikov, V. and Matthay, M. (2009). Allogeneic Human Mesenchymal Stem Cells for Treatment of E. Coli Endotoxin-induced Acute Lung Injury in the Ex Vivo Perfused Human Lung. *Proc. Nat. Acad. Sci. U.S.A.* 106: 16357–16362.
- Li, C.S. and Kuo, Y.M. (1992). Airborne Characterization of Fungi Indoors and Outdoors. *J. Aerosol Sci.* 23: S667–S670.
- Li, C.S. and Kuo, Y.M. (1993). Microbiological Indoor Air Quality in Subtropical Areas. *Environ. Int.* 19: 233–239.
- Li, C.S. and Wang, Y.C. (2003). Surface Germicidal Effects of Ozone for Microorganisms. *AIHA J.* 64: 533–537.
- Li, C.S., Hsu, C.W., Chua, K.Y. and Lin, R.H. (1996). Bioaerosol Characteristics in Daycare Centers. *J. Aerosol Sci.* 27: 653.
- Li, C.S., Hsu, C.W., Chua, K.Y. and Lin, R.H. (1997). House Dust Mite Allergens (*Der p* I and *Der p* V) within the Domestic Environments of Atopic and Control Children. *Arch. Environ. Health.* 52: 208–212.
- Naitou, S. and Takahara, H. (2008). Recent Developments in Food and Agricultural Uses of Ozone as an Antimicrobial Agent-food Packaging Film Sterilizing Machine Using Ozone. *Ozone Sci. Eng.* 30: 81–87.
- Najafi, M.B.H. and Khodaparast, M.H. (2009). Efficacy of Ozone to Reduce Microbial Populations in Date Fruits. *Food Control* 20: 27–30.
- Naydenov, A., Konova, P., Nikolov, P., Klingstedt, F., Kumar, N., Kovacheva, D., Stefanov, P., Stoyanova, R. and Mehandjiev, D. (2008). Decomposition of Ozone on Ag/SiO₂ Catalyst for Abatement of Waste Gases Emissions. *Catal. Today* 137: 471–474.
- Serra, R., Abrunhosa, L., Kozakiewicz, Z., Venâncio, A. and Lima, N. (2003). Use of Ozone to Reduce Molds in a cheese Ripening Room. J. Food Prot. 66: 2355–2358.
- Su, H.J., Wu, P.C. and Lin, C.Y. (2001a). Fungal Exposure of Children at Homes and Schools: A Health Perspective. *Arch. Environ. Health.* 56: 144–149.
- Su, H.J., Wu, P.C., Chen, H.L., Lee, F.C. and Lin, L.L. (2001b). Exposure Assessment of Indoor Allergens, Endotoxin and Airborne Fungi for Homes in Southern Taiwan. *Environ. Res.* 85: 135–144.
- Sundell, J. (2004). The History of Indoor Air Quality and Health. *Indoor Air* 14: 51–58.
- Taiwan Environmental Protection Administration (Taiwan EPA) (2005). Guidelines for Indoor Air Quality, Taipei, Taiwan, EPA.
- Takayama, M., Ebihara, K., Stryczewska, H., Ikegami, T.,

Gyoutoku, Y., Kubo, K., Tachibana, M. (2006). Ozone Generation by Dielectric Barrier Discharge for Soil Sterilization. *Thin Solid Films*. 506–507: 396–399.

- Tseng, C.C and Li, C.S. (2006). Ozone for Inactivation of Aerosolized Bacteriophages. *Aerosol Sci. Technol.* 40: 683–689.
- U.S. Environmental Protection Agency (US EPA) (1999), Ozone generators that are sold as air cleaners, Available from: http://www.epa.gov/iaq/pubs/ozonegen.html, last accessed 4 October 2011.
- Uhm, H.S., Hong, Y.F., Lee, H.Y. and Park, Y.H. (2009). Increase in the Ozone Decay Time in Acidic Ozone Water and its Effects on Sterilization of Biological Warfare Agents. J. Hazard. Mater. 168: 1595–1601.
- Wang, Y.C. (2003). Germicidal Effects of ozone on Bioaerosols, Master Thesis, Taipei, National Taiwan University.
- Weschler, C.J. (2006). Ozone's Impact on Public Health: Contributions from Indoor Exposures to Ozone and Products of Ozone-initiated Chemistry. *Environ. Health Perspect.* 114: 1489–1496.

- Whitney, E.A.S., Beatty, M.E., Taylor, T.H., Weyant, J.R., Sobel, J., Arduino, M.J. and Ashford, D.A. (2003). Inactivation of Bacillus Anthracis Spores. *Emerg. Infect. Dis.* 9: 623–627.
- Yoosook, W., Kongtip, P. and Chantanakul, S. (2009). Design and Development of Ultraviolet Germicidal Irradiation Device for Controlling Airborne Bacteria in the Workplace. J. Med. Assoc. Thai. 92: S142–S147.
- Yu, K.P., Lee, W.M., Lin, S.Y. and Huang, C.P. (2008). Removal of Bioaerosols by the Combination of a Photocatalytic Filter and Negative Air Ions. *J. Aerosol Sci.* 39: 377–392.
- Yu, Q., Zhao, M., Liu, Z., Zxang, X., Zheng, L., Chen, Y. and Gong, M. (2009). Catalytic Decomposition of Ozone in Ground Air by Manganese-based Monolith Catalysts. *Chin. J. Catal.* 30: 1–3.

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