

Indoor air purification by photocatalyst TiO₂ immobilized on an activated carbon filter installed in an air cleaner

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Abstract

The enhancement effect of using TiO₂ immobilized on activated carbon (TiO₂/AC) filter for removing indoor air pollutant at parts-per-billion (ppb) levels has been previously reported. To further evaluate the TiO₂/AC filter for practical application, it was installed in an air cleaner available in the commercial market and tested inside an environmental chamber. Nitrogen oxide (NO) and toluene were selected as target pollutant. Results showed that a higher removal efficiency of NO was achieved using shorter wavelength ultraviolet lamp than longer wavelength ultraviolet lamp. A higher NO removal was achieved using TiO₂/AC filter compared to TiO₂ filter only. The intermediate, NO₂, generated from the photodegradation of NO was also successfully suppressed from exiting the system using TiO₂/AC filter. A 25% higher of nitrogen oxides (NO_x) was achieved using TiO₂/AC filter compared to the TiO₂/AC only. The higher removal efficiency of using TiO₂/AC is owing to the large adsorption capacity provided by the activated carbon. The adsorbed NO is then transferred to the TiO₂ for photodegradation. The difference in toluene removal efficiency using TiO₂/AC filter compared to the TiO₂ filter is even more significant. © 2004 Elsevier Ltd. All rights reserved.

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1. Introduction

Indoor air quality has received immense attention in the early 1990s. This is because studies (Jones, 1999; Wallace, 1991; Niemala and Vaino, 1985) showed that the level of pollutants in indoor environment is actually higher than in outdoor environment. In addition, people generally spend more than 80% of their time in indoors, which contributes a higher risk from inhalation of pollutants than outdoors (Robinson and Nelson, 1995). In 1995, USEPA identified indoor air pollution is one of the top environmental risk (USEPA, 1995).

In general, three methods are suggested to improve indoor air quality, namely source control, increase ventilation and air cleaning. Source control is often ungovernable and unavoidable in metropolis such as Hong Kong. For instance,

vehicular exhaust from nearby traffic (Li et al., 2001), building materials (Hines et al., 1993) and the use of cooking utensils (Lee et al., 2001, 2002) were inevitable sources of indoor air pollutants. Increase ventilation might even transport more pollutants from outdoor environment (Jones, 1999). Thus, air cleaning remains to be the most feasible option to improve indoor air quality.

Recently, photocatalytic oxidation has shown to be a promising and effective technology for pollution control (Yu et al., 2002; Fujishima et al., 2000; Obuchi et al., 1999; Peral et al., 1997; Hoffmann et al., 1995). Unlike traditional pollution control method such as adsorption which merely transfers pollutant from gaseous phase to solid phase, PCO actually oxidizes pollutants to CO₂ and H₂O. However, studies (Luo and Ollis, 1996; Kim and Hong, 2002) showed that the rate of PCO decreased with decreasing pollutant concentration. In addition, at high humidity levels, water vapor competed with TiO₂ for adsorption sites which further decreased the rate of PCO (Ao et al., 2003a,b). Thus,

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in indoor environment where the pollutant concentration is only parts-per-billion (ppb) levels and under high humidity levels, the rate of PCO is rather low.

To improve this situation, Shiraishi et al. (2003) used an adsorbent to adsorb the pollutants to increase the pollutant concentration from the diluted air stream. The pollutant concentrated on the adsorbent was then desorbed upon heating, followed by TiO₂ photocatalytic oxidation. The HCHO removal efficiency was three times higher using the combination of adsorption/desorption and PCO compared to PCO only. Our group has also developed an activated carbon filter immobilized with TiO₂ (TiO₂/AC) to overcome the competition effect between the water vapor and the pollutant at ppb level for indoor air purification (Ao and Lee, 2003). The removal efficiency of TiO₂/AC was seven times higher than TiO₂ only under high humidity levels for multiple indoor air pollutants removal (Ao and Lee, 2004).

The aim of this study is to further investigate the feasibility of TiO₂/AC for practical indoor air purification. The TiO₂/AC filter was installed in an air cleaner available in the commercial market. The air cleaner was then evaluated inside an environmental chamber. Nitrogen oxide (NO) and toluene were selected as target pollutant as these pollutants are commonly found in indoor air. Sensitive analyses were conducted using different UV lamps and filter combinations. To the best of our knowledge, no detailed investigation has been reported on the evaluation of TiO₂ immobilized on an activated carbon filter installed in an air cleaner.

2. Experimental

2.1. Reagents and catalyst preparation

NO (BOC gas) was used as reactant gas and acquired from compressed gas cylinder at a concentration of 50 ppm ± 2% with nitrogen as balanced gas with traceable National Institute of Standards and Technology (NIST) standard. Toluene (Riedel-de Haen, reagent grade, 99.7%) was acquired from liquid phase.

TiO₂ (Degussa P-25) was used as a photocatalyst without any pretreatment. Activated carbon filter was purchased from a local company. The Brunauer–Emmett–Teller (BET) surface was 1115 m²/g. Using the desorption branch of the isotherm and the Barrett–Joyner–Helenda (BJH) formula, the average pore size was estimated to be 3.4 nm. The preparation of the TiO₂ filter and TiO₂/AC filter was described in elsewhere (Ao and Lee, 2003). Water suspension with 5% of TiO₂ was coated on a glass fiber filter (Whatman) as a supporting substrate (TiO₂ filter). It was then calcinated at 120 °C for 1 h with a temperature gradient of 5.5 °C/min. The same procedure was followed for TiO₂ loaded on activated carbon filter (TiO₂/AC), except an activated carbon filter (23.085 g ± 0.5%) was used instead of a glass fiber filter. The surface area of the glass fiber filter was identical to the activated carbon, which was 300 cm² used in this study.

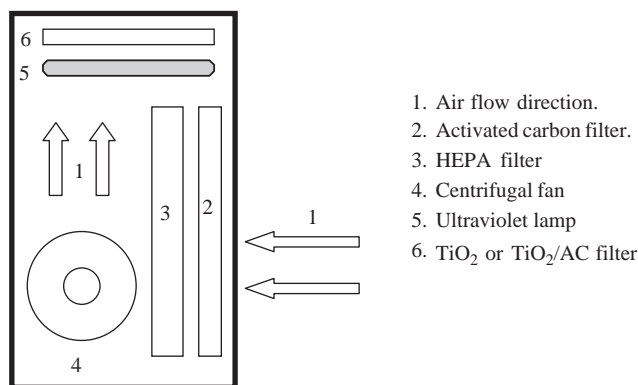


Fig. 1. Schematic diagram of the air cleaner.

The amount of TiO₂ imposed was determined by the weight difference before and after the coating procedure. In all experiments, the weight of TiO₂ imposed is 1.17 g ± 5%.

2.2. Environmental chamber

An electropolished stainless steel environmental chamber with a volume of 2.38 m³ was used in this study (Lee et al., 2003; Kwok et al., 2003; Lam et al., 2001). The temperature and relative humidity in all studies were 24 ± 1 °C and 70% ± 5%, respectively. Four mixing fans were installed at the bottom corners of the chamber to ensure adequate mixing of air. The temperature and relative humidity were continuously monitored by portable Q-Trak monitor (Model 8550, TSI, MN, USA). After each test, the chamber was cleaned by scrubbing the inner surfaces with deionized and distilled water. The chamber was then conditioned by purging zero air (Thermo Environmental Inc. Model 111) and conditioned to 25 °C and 70% relative humidity for at least 24 h prior to testing. The chamber background level was measured before each experiment.

2.3. Air cleaner

As shown in Fig. 1, the air cleaner is equipped with a high-efficiency particulate air filter (HEPA) and an activated carbon filter (AC). Illumination was provided by a 6 W ultra violet germicidal lamp (Sankyo Denki, Japan). The wavelength of the germicidal lamp ranged from 200 to 300 nm with the maximum light intensity at 254 nm (denoted as UVC). The ultra violet germicidal lamp can be interchanged by a 6 W black lamp (Sankyo Denki, Japan), depending on the experimental settings. The wavelength of the black lamp ranged from 300 to 400 nm with a maximum light intensity at 365 nm (denoted as UVA). A centrifugal fan is used to draw air at the front of the air cleaner and exhausted on the top, as indicated by the airflow directions in Fig. 1. The flowrate used in all study was 5.1 m³/min. The TiO₂ filter or the TiO₂/AC filter was mounted 1 cm on top of the UV lamp. The UV intensity measured (UVP radiometer, model

UVX) at the surface of the TiO_2 filter was 1.49 mW/cm^2 at 365 nm and 2.76 mW/cm^2 at 254 nm using the UVA lamp and UVC lamp, respectively.

2.4. Sampling and analysis

The air cleaner was placed inside the environmental chamber under static condition. NO and toluene were injected inside the chamber via a stainless sampling port. Mixing was allowed for 30 min and sample of the initial concentrations were collected via the sampling port with Teflon tubing at the center of the environmental chamber at 0.6 m above floor into a Teldar sampling bag (SKC). The concentration of NO was measured by a Chemiluminescence NO analyzer (Thermo Environmental Instruments Inc. Model 42c), which monitors NO, NO_2 , and NO_x at a sampling rate of 0.7 l/min. The concentration of toluene was measured by a non-methane hydrocarbon (NMHC) analyzer equipped with a FID detector (Thermo Environmental Instruments Inc. Model 55) at a sampling rate of 1 l/min. The air cleaner was turned on from an external circuit equipped outside the environmental chamber. Samples were then taken at desired time intervals.

3. Results and discussion

3.1. Blank test

Prior to the evaluation of the air cleaner, a blank test was conducted for NO and toluene. An initial 200 ppb NO concentration was achieved by controlling the amount of 50 ppm NO injected from the standard gas. Thirty minutes was allowed for mixing and the initial concentration was collected and analyzed. In all experiments, the initial NO and toluene concentration was controlled to be 200 ± 5 ppb and 2.15 ± 0.05 ppm, respectively. At time intervals 30, 60 and 120 min, samples were collected. As shown in Fig. 2(a), the initial NO concentration was similar to the NO concentration at 120 min. Similar result was obtained for toluene, as shown in Fig. 2(b). Thus, the environmental chamber was suitable for the testing of air cleaner without pollutant removal due to chamber leakage. Photolysis study was also conducted for UVA and UVC lamp. No removal of NO was observed using UVA lamp, whereas 20 ppb of NO removal was observed using UVC lamp.

Apart from the experiments with the indication of AC+HEPA setting, all the experiments were conducted without the original AC filter from the air cleaner. The HEPA filter was remained to maintain a constant flow for all experiments.

3.2. NO removal of the air cleaner

The original setting (AC+HEPA) of the air cleaner was evaluated to identify the removal of NO. This evaluation

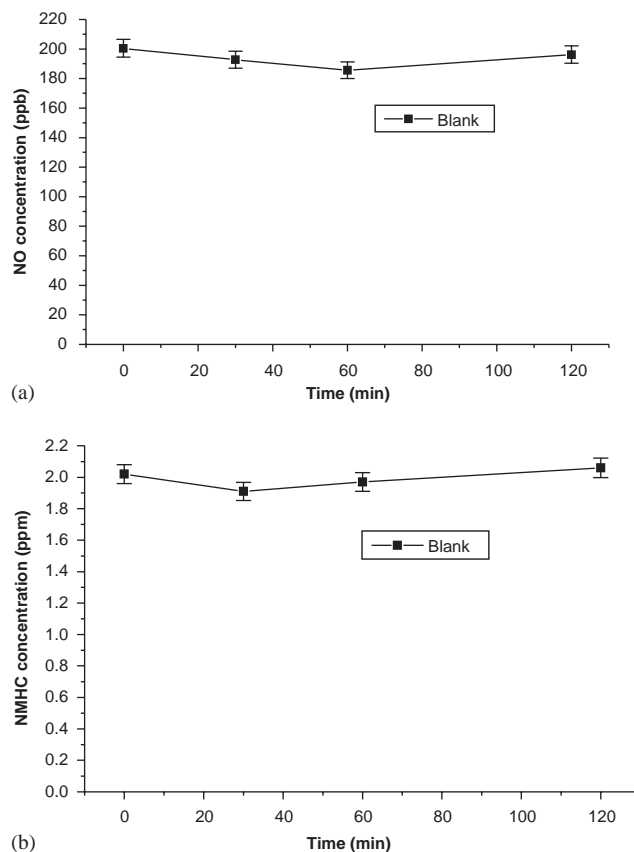


Fig. 2. Leakage test of the environmental chamber using (a) NO; (b) toluene.

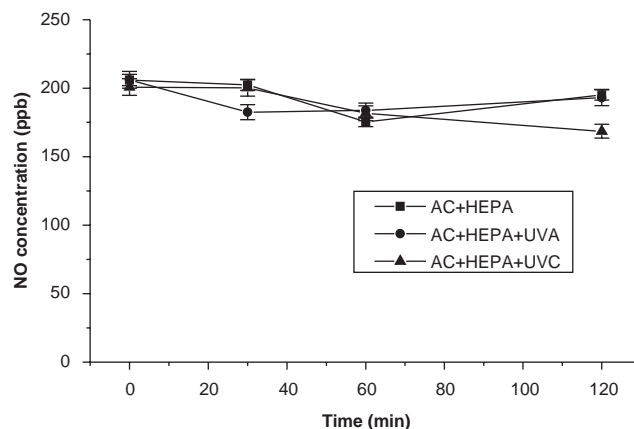


Fig. 3. NO removal using the air cleaner settings.

is vital as to truly investigate the removal efficiency of TiO_2 filter and TiO_2/AC filter mounted on the air cleaner despite the removal efficiency of the AC+HEPA. Fig. 3 shows three settings of the air cleaner namely AC+HEPA, AC+HEPA+UVA and AC+HEPA+UVC. The NO concentration decreased from 200 to 175 ppb at 60 min using AC+HEPA. However, the NO concentration increased back

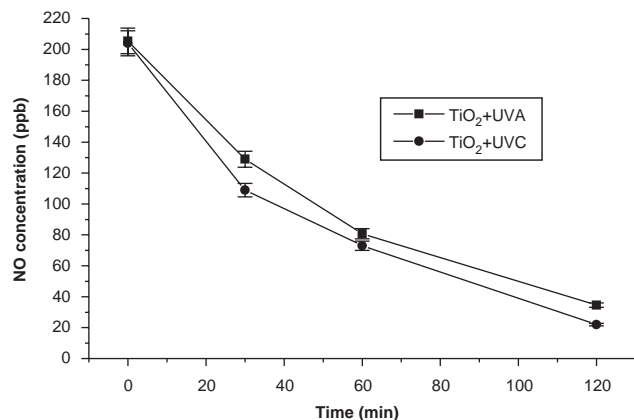


Fig. 4. NO removal using TiO₂ filter with UVA lamp and UVC lamp.

to around the initial concentration at 120 min. Presumably, the initial decrease of the NO concentration is due to the NO adsorption capacity of the activated carbon filter (Teng and Suuberg, 1993). The increase in NO concentration is probably due to the release of NO from the AC filter. Desorption of pollutant was also reported from AC filter (Schleibinger and Ruden, 1999). Similar result was observed for AC+HEPA+UVA setting. The NO concentration decreased at 60 min but increased to the initial concentration at 120 min. The use of UVA lamp was not able to remove NO, as reported in other studies (Devahasdin et al., 2003). Using UVC lamp instead of UVA lamp, however, a decrease of 25 ppb NO was observed at 120 min. Since the wavelength of the UVC lamp ranged from 200 to 300 nm, it included the commencement (Flory and Johnston, 1935) of the NO adsorption band and thus a reduction in NO concentration was observed.

3.3. NO removal of the air cleaner with TiO₂ filter using UVA and UVC lamp

Fig. 4 shows the photodegradation of NO using UVA lamp and UVC lamp. The time profile of NO photodegradation is identical using UVA lamp and UVC lamp. However, a higher NO removal was observed for UVC lamp. The NO concentration at 120 min using UVA lamp is 34.6 ppb, which is 12.6 ppb higher than using UVC lamp. This is probably due to a higher UV intensity was applied using UVC lamp. The measured UV intensity of UVC lamp (2.76 mW/cm²) was nearly double than that using UVA lamp (1.49 mW/cm²). Thus, at a higher UV intensity, more hydroxyl radicals were generated (Li et al., 1998; Kim and Hong, 2002) to react with NO and a higher NO removal was achieved. In addition, study (Matthews and McEvoy, 1992) also showed that TiO₂ particles adsorbed light more strongly for shorter wavelength than longer wavelength and reduced time participating in energy wasting recombination reactions.

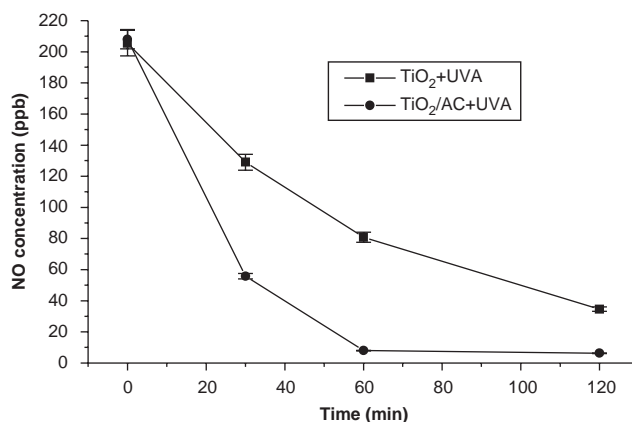


Fig. 5. NO removal using TiO₂ filter and TiO₂/AC filter.

3.4. NO removal of the air cleaner with TiO₂ filter and TiO₂/AC filter

Fig. 5 shows the photodegradation of NO using TiO₂ filter and TiO₂/AC filter. The NO concentration at 120 min was 34.6 and 6.3 ppb using TiO₂ and TiO₂/AC, respectively. Also shown in the same figure, the NO concentration reached a steady state at 60 min whereas the NO concentration using TiO₂ filter was still decreasing. Under such a small residence time and low NO concentration, the probability of NO contact with the hydroxyl radicals is rather low (Shiraishi et al., 2003). The use of activated carbon is to increase the adsorption of NO. The adsorbed NO is then transferred to TiO₂ for photodegradation. Study using TiO₂/AC in the photoreactor also showed that the effect of using activated carbon filter as supporting substrate is more significant at a lower residence time. At a high residence time, the pollutant diffusion rate from the gaseous phase to TiO₂ is similar to the pollutant diffusion rate from the activated carbon filter to TiO₂ (Ao and Lee, 2003). However, when such a high residence time is not applicable such as the air cleaner, the enhancement effect of using the activated carbon as the supporting substrate for TiO₂ is more significant than in the photoreactor.

Another possible reason for the higher NO removal using TiO₂/AC is the high relative humidity level used in this study. Water vapor competed with NO for adsorption sites on TiO₂ which reduced the NO removal rate (Ao et al., 2003a). Studies also showed that the oxidation rate decreased with increasing humidity levels for toluene (Cao et al., 1999) acetone (Kim and Hong, 2002), and 1-Butene (Cao et al., 2000). The above authors elucidated that water molecules competed with pollutants for adsorption sites on TiO₂ surfaces. Study also showed that the TiO₂ surface is full of hydroxyl groups which adsorbed water via hydrogen bond. Benzene and toluene, for instance, adsorbed stronger on dehydroxylated surface than hydroxyl surface (Nagao and Suda, 1989). Although at high humidity levels, water competed with NO for adsorption sites on TiO₂ and activated carbon, the large adsorption capacity of the activated carbon still able to

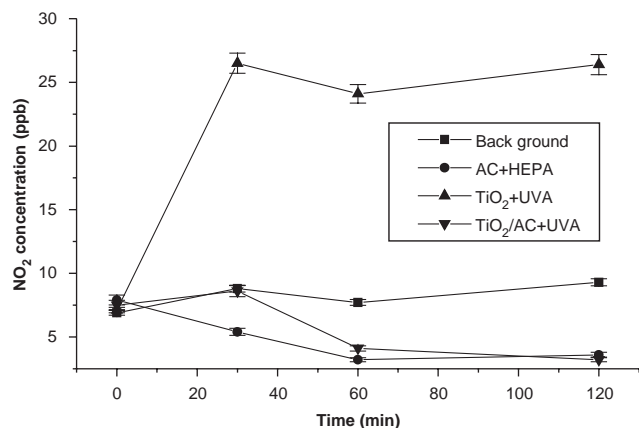


Fig. 6. Intermediate generated from the photodegradation of NO under different experimental conditions.

adsorb NO (Ao and Lee, 2004) because its surface area is 24 times larger than TiO₂. The adsorbed NO was then diffused to the TiO₂ for photodegradation. Using TiO₂ only for indoor air purification at ppb level pollutant concentration, NO competed with water for adsorption sites and the probability of contacting the hydroxyl radicals is low. Thus, the application of TiO₂/AC filter showed a promising and efficient method for indoor air purification.

3.5. Intermediate generated from the photodegradation of NO

Studies (Hashimoto et al., 2001; Nakamura et al., 2000; Dalton et al., 2002) showed that NO₂ is the intermediate generated from the photodegradation of NO, as shown in Eqs. (1) and (2):



In this study, NO₂ was also observed as the intermediate from the photodegradation of NO. As shown in Fig. 6, around 6 ppb of NO₂ was observed as the background concentration inside the environmental chamber. The NO₂ concentration decreased from 7.9 to 3.6 ppb at 120 min for the AC+HEPA setting. The decrease in NO₂ concentration is probably due to the adsorption of NO₂ on the AC filter (Rubel and Stencel, 1996). However, using TiO₂ filter with UVA lamp, the NO₂ concentration increased from 7 to 26 ppb. Using TiO₂/AC filter, the NO₂ concentration increased slightly from 7.1 to 9.2 ppb at 30 min. The NO₂ concentration then decreased to 3.2 ppb at 120 min. The large discrepancy of NO₂ concentration generated is due to the difference in using TiO₂ and TiO₂/AC. According to Eq. (2), NO₂ generated from the photodegradation of NO would further photo-oxidized to HNO₃. The residence time of the air cleaner provided was too low and NO₂ exited the air cleaner without further photodegradation. Using TiO₂/AC,

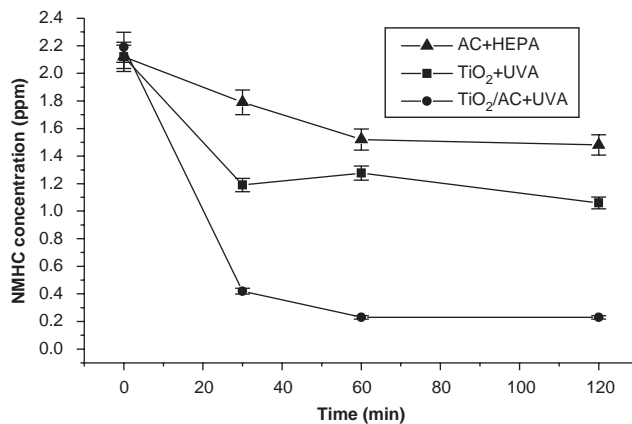


Fig. 7. Toluene removal by the air cleaner with AC+HEPA, TiO₂ filter and TiO₂/AC filter.

however, the activated carbon provided another adsorption site for NO₂. Similar to NO, NO₂ adsorbed on AC is then diffused to TiO₂ and photo-oxidized to HNO₃. Study (Torimoto et al., 1996) also showed that the combination of TiO₂ with adsorbent not only increased the propylamide removal rate but also reduced the amount of intermediate exited the system.

To accurately and truly evaluate the removal efficiency of NO using TiO₂ and TiO₂/AC filter mounted in the air cleaner, the following equation is used:

$$\text{NO}_x \text{ removal efficiency} = \frac{\text{Initial NO}_x \text{ conc.} - \text{final NO}_x \text{ conc.}}{\text{Initial NO}_x \text{ conc.}} \times 100\%, \quad (3)$$

where initial NO_x concentration is the initial NO and NO₂ concentration inside the environmental chamber and the final NO_x concentration is the NO and NO₂ concentration inside the environmental chamber at 120 min. The use of NO_x rather than merely NO for the pollutant removal efficiency evaluation because NO₂ generated from the photodegradation of NO is also a common indoor air pollutant (Jones, 1999). By applying Eq. (3), the NO_x removal efficiency using TiO₂ filter and TiO₂/AC filter is 70.5% and 95.4%, respectively. Thus, the use of TiO₂/AC not only increased the NO (target pollutant) removal rate but also reduced the generation of NO₂(intermediate).

3.6. Toluene removal of the air cleaner with TiO₂ filter and TiO₂/AC filter

Fig. 7 shows the photodegradation of toluene using the AC+HEPA, TiO₂ filter and TiO₂/AC filter with the UVA lamp. The use of AC+HEPA setting of the air cleaner decreased the NMHC concentration from 2.12 to 1.48 ppm. This is probably due to adsorption of toluene on the activated carbon filter, as reported in other studies (Urano et al., 1982; VanOsdell et al., 1996). However, the toluene removal is lower compared to TiO₂+UVA and TiO₂/AC+UVA. At a

Table 1
NO, NO_x and NMHC removal efficiency using TiO₂ filter and TiO₂/AC filter

Pollutant	TiO ₂ filter removal efficiency (%)	TiO ₂ /AC filter removal efficiency (%)
NO (ppb)	83.2	97.0
NO _x (ppb)	70.5	95.4
NMHC (ppm)	50.0	89.5

relative humidity higher than 60%, capillary condensation of water vapor occurred within the pores of activated carbon and making them unavailable for organic vapor adsorption (Cal et al., 1996; Khan and Ghoshal, 2000). Since the experiment was conducted at a relative humidity of 70%, the competition of adsorption sites on activated carbon between water vapor and toluene reduced its removal efficiency.

Similar to the photodegradation of NO, the use of TiO₂/AC achieved a lower NMHC concentration. The NMHC concentration was 1.06 and 0.23 ppm using the TiO₂ filter and TiO₂/AC filter, respectively. The toluene removal efficiency difference between the TiO₂ filter and the TiO₂/AC filter was larger than NO removal efficiency. As shown in Table 1, the difference in NMHC removal efficiency is 49.5% whereas only 13.8% in NO removal efficiency is observed. Similar results were reported in elsewhere (Ao and Lee, 2003). The high NO removal efficiency by the TiO₂ filter might hinder the enhancement effect of the TiO₂/AC filter. Using a less photoreactive compound such as toluene, the enhancement effect of the TiO₂/AC filter is more significant, as shown in this study. However, under the presence of other pollutants, such as hydrocarbons, the removal rate of NO is reduced due to competition of adsorption sites on TiO₂ (Ao and Lee, 2004). Thus, the difference in NO removal efficiency between using TiO₂ filter and TiO₂/AC filter will be higher if other pollutants are present.

4. Conclusion

A commercial air cleaner installed with a TiO₂ filter and a TiO₂/AC filter was evaluated inside an environmental chamber. Leak test showed that no pollutant removal was due to chamber leakage. The original setting (AC+HEPA) of the air cleaner showed no NO and little toluene removal. The use of TiO₂ filter removed 83.2% of NO but generated 12.9% of NO₂. Using TiO₂/AC, the NO removal efficiency increased to 97% and the generation of NO₂ decreased to 1.6%. The use of TiO₂/AC not only increased the target pollutant removal efficiency but also reduced the amount of intermediate exiting the system. The removal efficiency of the TiO₂/AC filter compared to the TiO₂ filter is even higher when the target pollutant is less photoreactive, such as toluene, used in this study. Thus, under a low residence time, low pollutant concentration and high levels of relative humidity, the use

of TiO₂/AC filter showed a promising and efficient method for indoor air purification. This study also showed that the enhancement effect of the TiO₂/AC shown in the laboratory scale using the photoreactor is also verified by installing it into an air cleaner available in the commercial market.

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