



AALBORG UNIVERSITY
DENMARK

Aalborg Universitet

CLIMA 2016 - proceedings of the 12th REHVA World Congress

volume 7

Heiselberg, Per Kvols

Publication date:
2016

Document Version
Publisher's PDF, also known as Version of record

[Link to publication from Aalborg University](#)

Citation for published version (APA):
Heiselberg, P. K. (Ed.) (2016). *CLIMA 2016 - proceedings of the 12th REHVA World Congress: volume 7*.
Department of Civil Engineering, Aalborg University.

General rights

Copyright and moral rights for the publications made accessible in the public portal are retained by the authors and/or other copyright owners and it is a condition of accessing publications that users recognise and abide by the legal requirements associated with these rights.

- ? Users may download and print one copy of any publication from the public portal for the purpose of private study or research.
- ? You may not further distribute the material or use it for any profit-making activity or commercial gain
- ? You may freely distribute the URL identifying the publication in the public portal ?

Take down policy

If you believe that this document breaches copyright please contact us at vbn@aub.aau.dk providing details, and we will remove access to the work immediately and investigate your claim.

Green Aspects of the Photocatalytic Oxidation (PCO) Method – Reference to the UV Turbo System

Karna Dahal^{#1} and Jouni Laurikainen^{*2}

[#]*Department of Environmental Science, University of Helsinki
Viikinkaari 2a, 00014, Helsinki, Finland*

¹karna.dahal@helsinki.fi

^{*}*Jeven Oy*

Malminkaari 23B 00700, Helsinki, Finland

²jouni.laurikainen@jeven.fi

Abstract

Cooking activities generate dirty exhaust air containing grease particles and odorous compounds. The ultraviolet (UV) turbo system represents an appropriate clean technology solution for purifying kitchen exhaust air. This advanced technique is based on the photocatalytic oxidation (PCO) reaction using ozone- (O_3) free UV light and titanium dioxide (TiO_2) as the catalyst. We tested the UV/ TiO_2 technique in the laboratory and a real kitchen environment to confirm the efficiency of grease and odor removal. We carried out laboratory testing of the UV turbo system for grease removal by analyzing exhaust air samples collected from a laboratory ventilation duct. Hydrocarbon (C-H) bonds in samples were analyzed with a spectroscopy method using a Fourier Transform Infrared Spectroscopy (FTIR) machine. Odor removal tests for kitchen exhaust air were completed using an OMX-SRM handheld odor meter according to the methodology described in the manual. We carried out field tests for odor removal in two restaurants applying the same techniques used in laboratory tests. The PCO method used in the UV turbo system proved beneficial for grease and odor removal, removing 100% of grease spatters $>2 \mu m$ and more than 50% of grease vapors $<2 \mu m$. In addition, the system eliminates about 89% of odor produced in the kitchen. This technique does not consume any ozone, while TiO_2 acts as a green catalyst, and works efficiently with 24-W UV power carrying fire prevention capabilities. Thus, we found that the UV turbo system is green and safe.

Keywords: *ozone-free, TiO_2 , PCO, UV turbo*

1. Introduction

Unclean air generated from cooking may circulate through the entire heating, ventilation and air conditioning (HVAC) system in a building. Improving kitchen air quality certainly improves air quality within a house. Among the few techniques available to purify kitchen exhaust air, the photocatalytic oxidation (PCO) reaction represents an effective and viable technique. The PCO method enjoys wide application from commercial products to environmental solutions [4]. Recently, this method proved quite attractive for air ventilation and indoor air-cleaning systems. As a technique, it not

only reduces grease, but also removes odors [8]. The photocatalyst used in the PCO reaction carries a self-cleaning property in rain without any harmful effects [4].

Kitchens represent the primary culprit for degrading air quality in buildings. Cooking activities generate dirty exhaust air which contain grease particles and odorous compounds. Grease particles deposit on kitchen walls, ventilation ducts, kitchen equipment, and building roofs. Grease is quite a sticky substance composed of different sizes of solid and liquid particles, grease and water vapors, and different non-condensable gases [3]. Grease particulates vary in size from 0.01 to 100 μm , with grease vapors limited to less than 0.01 μm [6]. Grease vapors are condensable, collecting in exhaust ducts and hoods [2]. Both grease vapors and particulates prove difficult to separate from kitchen fumes. Both accumulate in exhaust ducts and on the roofs of buildings, ultimately damaging fans and other equipment in ventilation systems. This accumulation may cause large fires in buildings causing damage to building structures and human beings. In addition, grease particulates larger than 10 to 20 μm become too heavy to be airborne and can drop from the airflow [6]. Unpleasant cooking smells can, however, spread throughout the surrounding environment [2].

Kitchen exhaust air should be filtered properly before extracting it to the outer atmosphere. In addition, efficient heat recovery requires relatively clean air without grease sticking to the recovery fins. Currently, all of the available grease removal systems consume high amounts of energy, generate harmful ozone gases, and inefficiently remove grease. However, some kitchen ventilation manufacturing companies are developing greener and more sustainable products. One such product consists of the UV turbo system, developed by Jevon Oy in Finland. Relying on the PCO method, which is green and can be used in kitchen air-cleaning systems, renders the UV turbo system greener than previous systems.

2. Background

Photocatalytic oxidation (PCO) is a reaction method accelerated by photocatalysts in the presence of light. Photocatalysts typically consist of semiconductor solids activated by UV light spectrums [400 nm–10 nm]. Pure or doped metal oxide semiconductors, such as Titanium dioxide (TiO_2), ZnO, CdS, and Fe (III)-doped (TiO_2), are commonly used as photocatalysts in the PCO method [12]. In the early 1970s, the Honda–Fujishima effect was discovered, where photo-induced water cleavages on TiO_2 electrodes [12]. TiO_2 is only photocatalyst used in commercial applications because it yields the most efficient photoactivity, the highest stability, and the lowest cost [4]. After the discovery of the Honda–Fujishima effect, studies in the 1980s focused on photocatalysis with powdered TiO_2 and moved to TiO_2 film- and surface-coating photocatalysis under weak UV light in the early 1990s [4]. A surface-coating photoreaction of TiO_2 resulted in multiple applications in building materials and many other areas [4]. TiO_2 features a self-cleaning function due to its angular surface tension properties [4]. TiO_2 -coated materials used in rainfall-exposed areas, such as exterior tiles, glass, windows, aluminum walls, and PVS fabrics, show quite effective

self-cleaning functions [4]. TiO₂ consists of three phases: anatase, rutile, and brookite, among which anatase exhibits the best photo activities [7].

In a Master's thesis project, we studied the effects of the photocatalytic reaction in the removal of grease from kitchen exhaust air through the combined application of UVC (100–280 nm) light at 254 nm and the anatase phase of TiO₂. However, as a part of product development, we completed further studies to upgrade the TurboSwing system (UV turbo) for the same purpose.

The PCO reaction occurs when UV light strikes TiO₂-coated surfaces. The conduction band of TiO₂ is excited by UV photons that create electrons and holes. Charges are transferred between electron-hole pairs and adsorbed pollutants, such as grease and odorous compounds, on the surface of TiO₂ [12]. The PCO reaction occurs as described below.

When adequate light strikes TiO₂, it breaks down into electrons and holes:

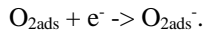


Then, oxidation and reduction reactions occur with air and water molecules present in the exhaust air.

The oxidation reaction is as follows:



The reduction reaction is written as



Hydroxyl radical (OH[·]) is the primary oxidant destroying organic compounds and in the presence of oxygen can prevent the re-combination of electron-hole pairs [12].

The complete reaction produces CO₂ and H₂O as the final products:



The efficiency of PCO depends on the photocatalysts, humidity, reactor type, and light sources [12].

Similarly, we also studied the odor removal capacity of the PCO method for the UV turbo system. These activities aimed to assist the development of a green UV turbo system—that is, a system which is energy efficient, safe, economical, and environmentally friendly. “UV Turbo” represents the revised name for the TurboSwing system after we implemented an advanced grease filtration technique based on the PCO reaction. UV Turbo initially performs grease filtration based on the rotational speed of the mechanically round, motor-operated grease filter, and, then, the PCO method. The mechanically round grease filter eliminates grease particles and vapors larger than 2 μm [5]. The UV light used in the advanced technique is ozone free, which prevents the

harmful effects of ozone. Many HVAC companies employ the ozonolysis technique in their odor-removing equipment, yet this technique is harmful and hostile to the environment. Ozone is a strong, oxidizing, harmful gas, where the uptake of >0.1 ppm ozone for periods of 8 hours or more can carry severe health problems [9]. Short-term exposure of ozone >0.3 ppm can also cause health-related issues [9]. The European Union's air quality directive sets threshold values for long-term ozone exposure at a mean concentration of $120 \mu\text{g}/\text{m}^3$ for 8 hours with a short-term (1 hour) exposure alert set to $240 \mu\text{g}/\text{m}^3$ [9]. Short-term exposure causes breathing difficulties, eye irritations, and itching similar to allergies, while long-term exposure leads to asthma, lung cancer, skin cancer, eye problems, and psychological disorders [11].

3. Methods

We tested the PCO (UV/TiO₂) technique in the UV turbo system both in the laboratory and in real kitchen environments in order to measure the efficiency of grease and odor removal. Odor removal experiments for kitchen exhaust air were carried out using an OMX-SRM handheld odor meter according to the procedures described in the manual. However, field observations for grease removal were also completed in a restaurant producing a large amount of grease. Field experiments for odor removal were carried out in two different restaurants using the same techniques used in the laboratory experiments.

3.1 Methods for Laboratory Work

Grease measurement. Laboratory tests of grease removal relied on analyzing exhaust air samples collected from a laboratory ventilation duct. We equipped the UV turbo with an 18-W ozone-free UV lamp and a rotating grease filter coated with anatase TiO₂ was connected to the ventilation duct in the laboratory. We used the sol-gel method for TiO₂ coatings. A series of experiments were completed for 1) the turbo alone, 2) the ozone-free UV lamp (18 W), and 3) the TiO₂-coated rotating plate and UV lamp (18 W) combined. Two different airflow rates (200 l/s) and (148 l/s) at two different ventilation duct heights (1 m and 9 m) were set up for the comparison studies. We placed sample cartridges with a suction pump at these points to collect exhaust air samples. Sigma sunflower oil from *Helianthus annuus* was heated to generate grease in the exhaust air. For each sample, 500 mL of oil was heated to 170°C for an hour. We collected air samples in filter papers, mixed them with tetrachloroethylene (TCE) solutions in beakers, shook them in an ultrasonic bath, and extracted the liquid. Hydrocarbon (C-H) bonds in the extracted samples were analyzed using the Fourier Transform Infrared Spectroscopy (FTIR) method to determine the grease concentrations. Each spectrum's web numbers of 2700 to 3100 cm^{-1} were

observed, and the best peaks—2888.5 to 2978.3 cm^{-1} —were taken into consideration for the calculation of the C-H bonds.

Odor measurement. We employed an OMX-SRM handheld odor meter to measure the odor of kitchen exhaust air. As in the grease measurements, 500 mL of sunflower oil was heated for 30 minutes to generate grease in a closed laboratory environment. We placed the odor meter at two different ventilation duct heights (1 m and 9 m). We completed numerous experiments using 1) the UV turbo system only, 2) an ozone-free UV lamp (18 W), and 3) TiO_2 -coated UV turbo walls combined with a UV lamp (18 W). We also tested the effect of an ozone-generating lamp for odor removal in similar experimental conditions in another kitchen air-cleaning system due to the difficulties of placing an ozone-generating lamp inside a UV turbo box. The odor concentrations provided by the meter were completely different from the odor sensed by humans [10].

3.2 Methods for Field Measurements

Grease measurement. We observed grease removal using the PCO technique in a restaurant operating 24 hours a day, 7 days week. TiO_2 -coated rectangular plates with a 24-W ozone-free UV lamp were installed in the kitchen hood inside the UV turbo system. We then observed the effects over three-week intervals for seven months. We installed an ozone-free UV lamp (24 W) without TiO_2 -coated plates in another UV turbo system and observed the effects of the UV lamp only for grease removal. We took photos of the turbo boxes, rectangular plates, installed lamps, and ventilation ducts used for analyses.

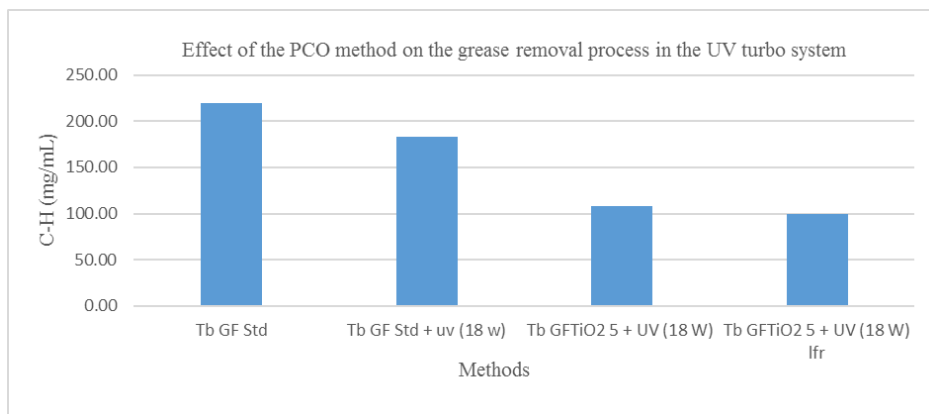
Odor measurement. We measured cooking odors in two different restaurants. The first restaurant was equipped with ozonators; we then measured cooking odors with and without the application of ozonators to determine the effects of ozone on odor reduction. We used the same handheld odor meter from the laboratory measurements to measure odors in the restaurants. The odor and ozone measurement point was situated 30 m away from the kitchen. In the second restaurant, the odor was measured without the application of UV/ TiO_2 initially. We then installed an ozone-free UV light (24 W) and TiO_2 -coated plates to observe the effect of the PCO reactions on odor reduction. With similar food menus and almost equivalent amounts of food cooked, we performed odor measurements during lunch between 10:45 and 14:30.

4. Results and Discussions

In this section, we present the results from both laboratory and field observations for grease and odor removal using the PCO technique.

4.1 Laboratory Results for Grease Filtration Using the PCO Method

Laboratory results revealed that 51% and 55% of grease vapors and particulates ($<2\ \mu\text{m}$) were reduced using the PCO (UV/TiO₂) method in the UV turbo system at an airflow rates of 200 l/s and 148 l/s, respectively. The UV lamp (18 W) at a power of 5-W UVC reduced grease vapors and particles by 14%. Only the surface area of the rotating grease filter was coated with TiO₂. If the walls of the UV turbo system were also coated with TiO₂, grease reduction would be increased by 51% at a higher airflow rate and by 55% at a lower airflow rate. Our results confirm that PCO works better at a lower than at a higher airflow rate.



¹ Tb = turbo, GF = grease filter, Std = standard, Lfr = low airflow rate, 5 = anatase TiO₂ phase]

Figure 1: Effect of the PCO method on the grease removal process in the UV turbo system

As shown in Figure 1, grease removal using a standard grease filter (GF Std.) was first tested without the PCO method. A standard grease filter refers to the grease filter used in the previous version of the UV turbo system (that is, the TurboSwing system). Then, we tested the grease removal capacity of GF Std with a 18-W UV lamp set to a 5-W UVC power. Finally, a TiO₂-coated rotating grease filter (GF) was

combined with a UV lamp. Our results verify that PCO works efficiently in the grease removal process.

4.2 Field Results for Grease Filtration Using the PCO Method

During field observations, we completed several observations of grease filtration using a UV lamp alone and a combined UV/TiO₂ system. During the observations over seven months, we clearly observed grease deposited on the walls of the turbo system, which was almost entirely removed by UV/TiO₂ over a five-month period. The turbo system was installed in one restaurant 3.5 years before our observations began, and was not cleaned by any means. Figures 2 and 3 illustrate the massive grease deposits in the box.

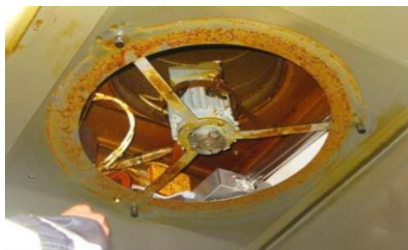


Figure 2: Turbo box and duct on 10 October 2014. Figure 3: Turbo box and duct on 13 March 2015.

As shown in the images above, the TiO₂-coated plates after 5 months were as clean as on the day it installed to the turbo box walls and rest turbo box walls nearby plates were also cleaned very well. The restaurant kitchen operated 24 hours a day 7 days a week. The effect of PCO was seen within three weeks of applying the UV/TiO₂ to the installed turbo box. Almost all grease deposited over a 3.5-year period was also removed along with any fresh grease produced from cooking.

*

4.3 Laboratory Results for Odor Removal Using the PCO Method

Odor concentration measurements from laboratory conditions revealed that the PCO technique is useful in odor removal. We reduced the sunflower oil odor produced in the laboratory by 26% at point 2 (1 m above the turbo box) and 52% at point 3 (9 m above the turbo box). The reduction range between these two points was large since odor cannot be measured properly given that odor formations and flows depend on atmospheric conditions such as temperature, humidity, and cooking

ingredients. During laboratory analyses, we also measured olive oil and sunflower oil odors. Odor reduction between these two types of oils differed by 10% at measurement point 2 under similar atmospheric conditions. It is clear that the odor concentration at point 3 was lower than that at point 2, because UV/TiO₂ reactions near the turbo chamber occurs at a higher frequency than above the ventilation duct. Odor removal using an ozone-generating lamp was a bit strange, because no oil odor was detected although we sensed a strong ozone odor during the experiments. The odor meter was not intended to measure the ozone odor, yet the realization of an ozone odor was strong and irritating. It is also possible that the oil odor was undermined by the ozone odor. However, we did not observe such issues with odor removal using the PCO method.

4.4 Field Results for Odor Removal Using the PCO Method

Practical observations of odor reduction using the PCO method were carried out at two different restaurants. Table 1 provides the ozone and odor concentrations during lunch (11:15–14:15) at the first restaurant, while the odor concentrations at the second restaurant using the PCO technique appear in Table 2.

Table 1: Measurements of odor and ozone levels at restaurant 1

Measurement time (11:15–14:15)			
Days	7 May 2015	8 May 2015	9 May 2015
Odor average (n/a)	4.1	3.8	5.2
Ozone average (ppm)	0.6	0.7	0

The cooking activities during the experimental observations were nearly identical each day. Table 1 clearly shows that large amounts of ozone were used in the kitchen ventilation. The measurement point was quite close to the ventilation outlet and ~0.6 to 0.7 ppm ozone dispersed from the outlet during the experiments. This ozone concentration is quite harmful to the surrounding environment (the limit for longer ozone exposure times stands at 0.1 ppm). Similarly, the generation of ozone and odor concentrations during the first, second, and third days suggest that ozone plays no significant effect on odor reduction since large amounts of ozone and odor were measured near the ventilation outlet.

In the second restaurant, we observed odor reductions of 29% to 89% during lunch (10:45–14:30) while preparing the same foods without UV/TiO₂ one week and with UV/TiO₂ during the following week. Odor reduction between the two Mondays under similar atmospheric conditions and with identical lunch menus reached 29% and

89% on Thursdays under similar conditions. The quantity of cooked food was roughly the same on both Mondays and Thursdays.

Table 2: Odor measurement with and without the PCO technique at restaurant 2

Days	without UV/TiO ₂		with UV/TiO ₂	
	Monday (27 July)	Thursday (30 July)	Monday (24 August)	Thursday (27 August)
Average	23.5	32.9	2.8	23.3
Humidity (rH %)	35.7–49.0	39.2–49.9	34.6–49.4	40.5–54.1
Temp (°C)	26.6–29.2	28.1–29.6	26.0–29.9	27.3–28.8
# of lunch portions	32	34	29	36

The reduction of cooking odors using the PCO (UV/TiO₂) was significant. We measured odor up to 3 m away from the turbo’s location in the kitchen. Thus, odor values after treatment were markedly higher. The odor reduction difference between Mondays and Thursdays was quite high, but generating the odor flow to the ventilation duct may not have relied on similar air streams because slight changes in kitchen environments result in different odor flows. For instance, if cooked foods were kept far away from the kitchen air-cleaning systems, the odor might flow in other directions instead of flowing to the ventilation ducts. If the kitchen environment featured multiple air movement disturbances, the kitchen exhaust flow also differed. Nevertheless, we completed our analyses assuming that the exhaust air flowing to the ventilation duct was similar during the test periods.

Our results verified that odor and grease reductions using the PCO technique are suitable for kitchen exhaust air-cleaning systems. However, PCO requires quality mechanical grease separation before the PCO process. A majority of grease particles >2 μm should be removed before application. Grease-deformed compounds after PCO flows with air, and some may remain on the plenum and duct. This can be easily cleaned mechanically. The UV turbo system used for grease filtration and odor removal also allows for the integration of a heat recovery system. The heat recovery system utilizes waste heat from exhaust air and improves the outgoing air quality and energy efficiency of the ventilation system. Particles flowing with air should be removed with a back filter before a) heat recovery and b) extraction from buildings. PCO works better at a lower airflow rate and kills bacteria and viruses present in kitchen ventilation systems [1]. TiO₂ coatings on larger surface areas, such as the walls, increases the efficiency of the UV turbo system. As such, a larger UV power is not necessary for the UV/TiO₂ reaction to occur. The UV turbo system currently operates with a 24-W UV lamp at a power of 8 W UVC, but an 18-W UV lamp set to a power of 5 W UVC allowed reactions to occur during laboratory testing. The total electric power consumed by the UV turbo system was just 54 W initially and 10 W

subsequently. Many kitchen air-cleaning systems consume 100 W to 200 W. This helps to reduce energy bills and provides financial incentives. Due to the longer-lasting effect of PCO for grease and odor removal in the UV turbo system, this also reduces the maintenance costs for the entire ventilation system including the UV turbo system. The UV turbo system is also energy efficient compared to other kitchen air-cleaning products.

5. Conclusions

Our analysis shows that the photocatalytic oxidation (PCO) reaction method is green and an appropriate technique for grease and odor removal. Furthermore, the PCO technique applied to the UV turbo system is analytically and practically verified as a useful method for kitchen air purification. It removes 100% of grease spatters $>2\ \mu\text{m}$ and more than 50% of grease vapors $<2\ \mu\text{m}$. It eliminates up to 89% of odor produced in the kitchen. This system works efficiently at 24-W UV power and carries fire prevention capabilities. As a technique, it is much greener than the photolysis or ozonolysis methods currently in use for grease and odor removal processes. It avoids harmful ozone gases in the grease and odor removal system and uses the green catalyst titanium dioxide (TiO_2) instead of ozone. PCO has improved the performance of the UV turbo system compared with previous versions, which operated with a rotating mechanical grease filter. PCO-based purified kitchen air extracted to the surrounding environment is cleaner than exhaust air purified using other techniques. Grease removal using the PCO method in the UV turbo system not only prevents possible fires due to grease deposition, but also prevents flume exposure to the ducts if a fire results from cooking. Therefore, PCO using UV/ TiO_2 is energy efficient, environmentally friendly, and economical. The efficiency of PCO applied to the UV turbo system may improve by extending the TiO_2 coatings to wider surfaces. We find that integrating the heat recovery system to the UV turbo system improves the air quality and energy efficiency of the system.

References

- [1] Bonetta, S.; Bonetta, S.; Motta, F.; Strini, A.; & Carraro, E. (2013). Photocatalytic bacterial inactivation by TiO_2 -coated surfaces. *AMB Express*, 3. doi:10.1186/2191-0855-3-59.
- [2] Dahal, K. (2014). Doria: Master's theses (technology). Available from National Library of Finland's publication site: Doria: https://www.doria.fi/bitstream/handle/10024/96430/Karna_Dahal_Master%20Thesis.pdf?sequence=2 [Accessed 15. 1 2016].
- [3] FCSI. (no date). Grease Extraction - Myth and Reality. Available at: https://c.ycmdn.com/sites/fcsi.site-ym.com/resource/resmgr/americas_quizes/fcsi_quiz_-_halton_grease_ex.pdf [Accessed 24. 1 2016].
- [4] Hashimoto, K.; Irie, H.; & Fujishima, A. (2006). TiO_2 Photocatalysis: A historical Overview and Future Prospects. *Japanese Journal of Applied Physics*, 44(12), 8269 -8285.

- [5] Jeven. (2014). TurboSwing:Jeven Oy. Available from Jeven Oy website: <http://www.jeven.fi/en/turboswing/> [Accessed 24. 1 2016].
- [6] Livchak, A.;Schrock, D.;Lehtimaki, M.;& Taipale, A. (2003). The facts. Mechanical Grease. Available at: <http://www.revacsystems.com/images/downloads/filterefficiency.pdf> [Accessed 14. 1 2016].
- [7] MO, S.-D.;& Ching, W. (1995). Electronic and optical properties of three phases of titanium dioxide: Rutile, anatase, and brookite. *Physical Review B*, 51, 13023 - 13032. doi:<http://dx.doi.org/10.1103/PhysRevB.51.13023>.
- [8] Nozawa, M.;Taniqawa, K.;Hosomi, M.;& Chikusa, E. (2001). Removal and decomposition of malodorants by using titanium dioxide photocatalyst supported on fiber activated carbon. *Water Sci Technol*, 44(9), 127-133.
- [9] Ozonotech. (2011). About Ozone: Ozonotech. Available from Ozonotech website: <https://www.ozonotech.com/about-ozone/ozone-safety> [Accessed 19. 1 2016].
- [10] Shinyei Technology. (2012). Handheld Odor Meter: Shinyei Technology. Available from Shinyei Technology website: <http://www.shinyei.co.jp/stc/eng/iaq/odor.html> [Accessed 18. 1 2016].
- [11] WHO. (2003). Health Aspects of Air Pollution with Particulate Matter, Ozone and Nitrogen Dioxide: WHO's Regional Office for Europe. Available from WHO's Europe website: http://www.euro.who.int/__data/assets/pdf_file/0005/112199/E79097.pdf [Accessed 24. 1 2016].
- [12] Zhao, J.;& Yang, X. (2003). Photocatalytic oxidation for indoor air purification: A literature review. *Building and Environment*, 38(5), 645-654. doi:[doi:10.1016/S0360-1323\(02\)00212-3](https://doi.org/10.1016/S0360-1323(02)00212-3).