

Development of a photocatalytic filter to control indoor air quality

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ABSTRACT

Background: The aim of this work was the development and characterization of a photocatalytic filter for the treatment of indoor air, characterized by a low pressure drop.

Methods: The filter (photocatalytic filter) was based on a polyester substrate additivated with active carbon (Carbotex 150-6), treated with a sol of titanium dioxide (Sol 121-AB; NextMaterials Ltd.) and illuminated with UV LEDs to induce photocatalytic activity.

Results: Tests showed that this filter, used in a suitable device for air circulation with a very low noise level, had the ability to block solid particulates, to photocatalytically oxidize a major fraction of volatile organic compounds (VOCs) and deactivate all of the bacteria blocked on the filter, in contrast to traditional commercial air filters on which the bacteria remain viable.

Conclusions: Activated charcoal filters treated with TiO₂ and illuminated by UV LEDs were found to be extremely effective in killing bacteria and effective in decreasing VOC and total suspended particulates (TSP).

Keywords: Bacteria, Filters, Photocatalysis, Titanium oxide, Total solid particulate, Volatile organic compounds

Introduction

The air quality in indoor environments (IAQ) may depend on many factors such as pollutants resulting from human activities (i.e., metabolic processes, tobacco smoke, cooking food, use of detergents and cleaning agents), construction materials and decor (emission of volatile organic compounds [VOCs]), air conditioning systems, combustion, and household and office appliances. All of these are added to external sources of pollution from the atmosphere, water and soil (1, 2). The danger from indoor pollutants is mainly determined by the fact that these substances are released in small spaces, which often have a poor air exchange with the outside, and thus tend to accumulate. Besides, although individual outdoor protection can be achieved by facial masks, indoor air usually relies on expensive and energy-intensive air-filtering devices (3).

IAQ control has thus become increasingly important in relation to complex forms of pollutants, in particular microbials

(4), VOCs (5) and total solid particulates (TSPs) (3), because over the last 2 decades, there have been an increasing number of instances where the occupants of buildings have complained of a general feeling of being unwell or of experiencing acute ill health and discomforting effects that appear to be linked to time spent in a building. This phenomenon is called sick building syndrome (SBS).

Bacteria are unicellular microorganisms belonging to the prokaryotes that exist as free-living organisms or as parasites and are spread almost everywhere. It is worthy of note that not all pests are really harmful to the host. There are in fact symbiotic bacteria, from which humans draw certain advantages (e.g., the vaginal flora). There are also commensal bacteria, that do not cause any damage, but also no benefit. Conversely, there are pathogens that are harmful to the body. For instance, in July 1976, at the Bellevue-Stratford Hotel in Philadelphia, a lung infection caused by the bacterium *Legionella pneumophila* struck 221 American Legionnaires who were attending an event (hence the name Legionella, as this form of pneumonia was previously unknown), causing the death of as many as 29 of them (6). The source of the infection was identified as bacterial contamination in the air conditioning system. This also enabled an understanding that the humidifiers of centralized systems are sources of bacterial endotoxins and suitable breeding grounds for bacteria such as *Legionella pneumophila* and some species of *Pseudomonas*, *Acinetobacter*, *Staphylococcus* and *Candida*.

It is also worthy of consideration that the smoke of tobacco releases nicotine, an alkaloid with strong psychoactive properties able to create addiction, and which can introduce

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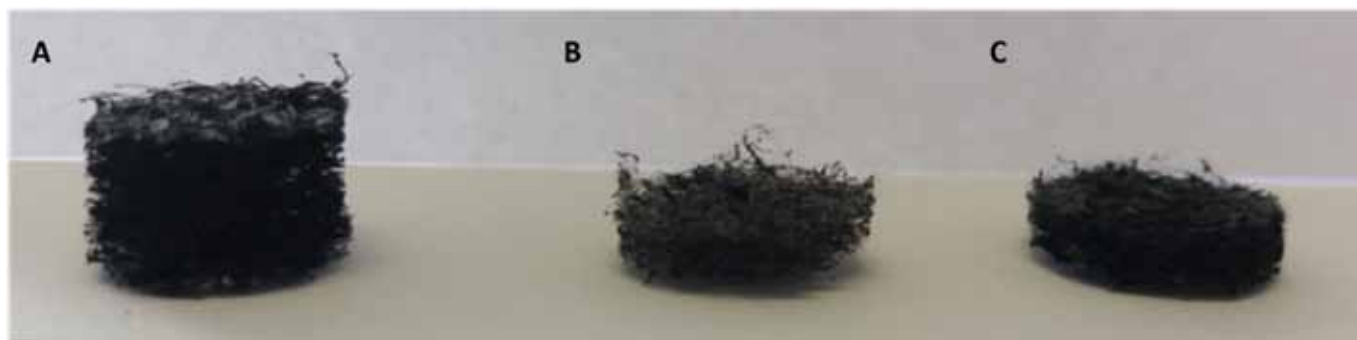


Fig. 1 - (A) Commercial filter, (B) Lot A photocatalytic filter and (C) Lot B photocatalytic filter.

carbon monoxide and carcinogenic substances into the indoor environment. Tobacco smoke is one of the leading causes of death: In Italy, 15% of all deaths can be directly attributed to smoking - i.e., about 80,000 people per year.

The damaging effects of TSP can be short term as in the case of inflammations and influences on chronic respiratory diseases, or they can cause chronic inflammation of the respiratory tract and increase the risk of lung cancer. They are considered the cause of 11% of deaths from chronic obstructive pulmonary disease, which is considered by the World Health Organization (WHO) to be the third cause of death in the world, with over 3 million deaths per year (5,876 deaths in only 15 Italian cities monitored per year).

In light of such issues, some air-cleaning devices have been developed and commercialized (3, 7, 8). Nevertheless, many shortcomings in these still exist. Specifically, even though traditional filters used in filtration systems developed so far are able to block solid particulates and bacteria, they are not able to eliminate individual sources of pollution or to reduce their emissions, because of the ability of bacteria to colonize the surfaces of such filters. Besides, traditional filters used in air filtration devices are not effective on VOCs. Moreover, high-efficiency particulate air (HEPA) filters and ultra-low particulate air (ULPA) filters, which are air filters designed to trap the vast majority of very small particulate contaminants from an air stream, give rise to high pressure drops and consequently to high energy consumption and ambient noise and require massive use of pumping systems (3).

Herein we proposed a new class of TiO_2 -treated UV LED activated charcoal filters and studied their antimicrobial activity on bacteria mechanically stuck on the filters, and their effectiveness in decreasing TSPs and VOCs. As such filters are characterized by a low pressure drop, they can be utilized in low-power, noiseless devices used for air ventilation.

Materials and methods

Filter prototyping

Photocatalytic filters have been developed and characterized, able not only to break down solid particles, but also, thanks to the use of photocatalysis (9-15), to oxidize VOCs and kill the bacteria that were mechanically locked onto the filter.

The photocatalytic filter was developed starting from an activated carbon filter with the following characteristics:

substrate polyester, active carbon 24%, active carbon surface area $40,000 \text{ m}^2/\text{m}^2$, flame retardancy FH2 (EN 60335-2-21), weight $150 \pm 15 \text{ g}/\text{m}^2$ (EN 29073-1), thickness $5.5 \pm 1.5 \text{ mm}$ (EN ISO 9073-2), and air permeability (pressure drop 25 Pa) $1,050 \pm 263 \text{ L}/\text{m}^2/\text{s}$ (EN ISO 9237).

The filters were treated using sol-gel technology to induce the precipitation of a titanium dioxide film (16-18). Sol 121-AB (NextMaterials Ltd.) with the following composition was used: water 95%, titanium dioxide 2%, functionalized silica 1%-2%, isopropanol 0-3%, acids $\approx 1\%$, dopant $\approx 1\%$, methanol $<0.5\%$, additives (silver salt) 1% and semitransparent.

LEDs used for the experiments were SMD 5050, wavelength 395 nm (purple), voltage 12 V, wattage 240 mW/each.

Antimicrobial activity

To assess in vitro the time-dependent reduction of a microbial population after exposure to different antimicrobial devices, we first checked the antibacterial effectiveness of the filters per se. Treated filters were grouped into 2 lots (Lot A and Lot B photocatalytic filters) and compared to a commercially sourced filter (Commercial filter) (Fig. 1).

Briefly, disc-shaped samples ($\varnothing = 15 \text{ mm}$, surface area = 1.75 cm^2) were cut from each kind of filter and transferred into a sterile 24-well plate (Fig. 2). An aliquot of gram-negative *Escherichia coli* (strain JM109, cat. 53323; American Type Culture Collection [ATCC], Manassas, VA, USA) was transferred onto a sterile lysogeny broth agar plate and incubated at 37°C for 60 minutes to allow the growth of bacterial colonies. After incubation, using a sterile inoculating loop, a single colony was transferred into a 50 mL polypropylene tube (Corning, Fisher Scientific, Rodano, Italy) filled with 5 mL of sterile LB liquid growth medium and incubated overnight (ON) at 37°C under shaking, to produce sufficient microbial suspension. The number of bacteria within the microbial suspension was roughly estimated by measuring the optical density (OD) of the suspension at $\lambda = 600 \text{ nm}$ ($\text{OD}_{600\text{nm}}$) by means of a Nanodrop 2000 spectrophotometer (Thermo Fisher Scientific, Segrate, Italy). Afterwards, the inoculum was prepared by diluting the microbial suspension in sterile LB to an $\text{OD}_{600\text{nm}} = 0.02$. Bacteria were next allowed to grow until they reached an $\text{OD}_{600\text{nm}} \approx 0.6$ (exponential growth phase). The inoculum was finally centrifuged for 10 minutes at $4,000 \text{ g}$, washed once in sterile deionized water (dH_2O), pelleted again, and then the cells were

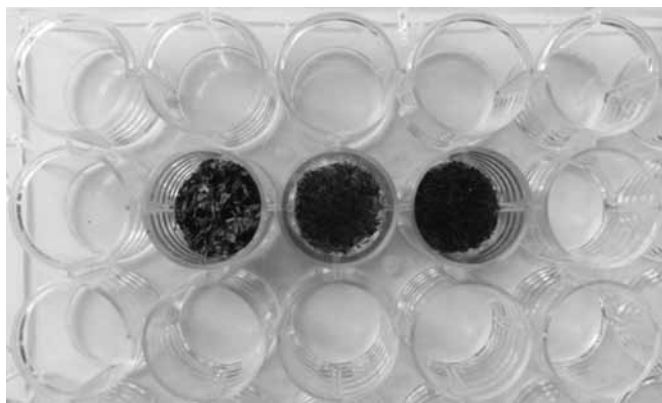


Fig. 2 - Samples from commercial and photocatalytic filters placed in a 24-well plate.

resuspended in dH_2O to give a bacterial concentration of $\approx 5 \times 10^8$ bacteria/mL.

Following a preincubation at 4°C , samples were brought into contact with the inoculum. The experiments were carried out according to the Assessment of Antimicrobial Activity Using a Time-Kill Procedure (ASTM E2315) (19) and Measurement of Antibacterial Activity on Plastics and Other Non-porous Surfaces (ISO 22196) (20) standards, slightly modified in terms of the bacterial inoculum (10^3 times higher bacterial load as compared with standards) and the temperature at which the experiments were performed (4°C instead of $35^\circ\text{C} \pm 1^\circ\text{C}$).

Briefly, each test sample (commercial filter, $n = 3$; photocatalytic filters, $n = 2$) was inoculated with 1 mL of the bacterial inoculum and incubated at 4°C for different durations (30 and 60 minutes). During the experiments, the photocatalytic filters underwent UV irradiation (Fig. 3). Bacteria inoculated in wells without any filter were used as positive controls (untreated bacteria).

At different time intervals, a $30\text{-}\mu\text{L}$ aliquot of bacterial suspension kept in contact with each sample was removed from the well, serially diluted in sterile dH_2O (1:10, 1:100, 1:1,000), plated onto P60 LBA plates and incubated ON at 37°C to allow the growth of bacterial colonies. After incubation, the number of viable bacteria was evaluated by counting the colony forming units (CFUs) grown on agar plates. The number of viable cells was standardized to the plate area (cm^2) and the dilution factor. Finally, data were normalized with respect to the number of viable cells in control samples at each time step, considering the number of untreated bacteria as 100%. Results were expressed as means \pm standard error of the mean (SEM).

Evaluation of TSP and VOC abatement

To evaluate TSP and VOC abatement, a square device closed on the sides was developed ad hoc. It consisted of:

- four cooling fans from a personal computer $92 \times 92 \times 25$ mm, 2,100 rpm brushless, 12 V, 0.18 A, air flow $1.16 \text{ m}^3/\text{min}$, air pressure $2.71 \text{ mm}_{\text{H}_2\text{O}}$, noise 27.8 dB(A), raised 20 mm with respect to the floor;
- four 15-cm LED strips (27 LEDs) positioned immediately above the fans;

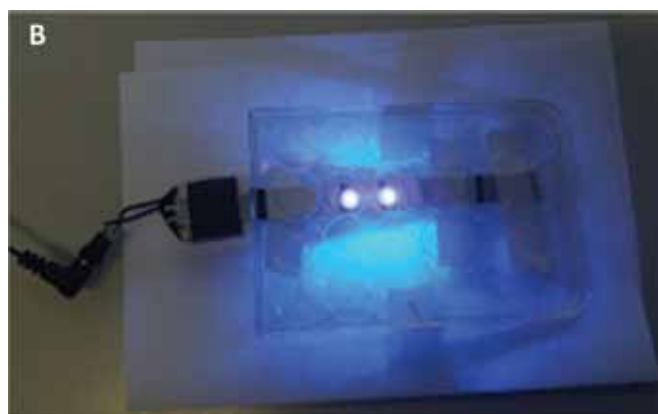
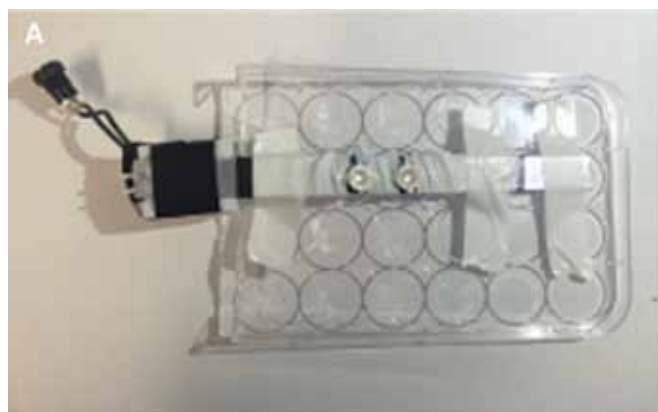


Fig. 3 - Picture of the UV LED lighting system: (A) before the experiments and (B) in use.

- one photocatalytic filter 184×184 mm raised 10 mm with respect to the LEDs.

Tests were performed in a Glove Box (100 L volume) polluted by one half medium nicotine content cigarette, able to rapidly generate a large amount of TSPs and VOCs.

The following testing procedure was used:

- ignition of the cigarette,
- immediate closure of the Glove Box,
- cigarette combustion,
- waiting for 16 minutes,
- switching on filter fan,
- monitoring of up to 120 minutes of the TSP content using an Aerocet 531 (Met One Instruments Inc.),
- monitoring for up to 120 minutes of VOC content using a DSIQ-PLUS-PPC (Gray Wolf Sensing Solutions).

The results obtained were then averaged ($n = 2$) and calculated as percentage decrease compared with the value at the time of the start of the fan (16 minutes).

Results and discussion

Antibacterial effectiveness

A low pressure drop filter was developed and characterized in this work. Due to its photocatalytic behavior (3-6),

such a photocatalytic filter was able to break down solid particles, oxidize VOCs and kill the bacteria that were mechanically blocked onto its surface.

The antibacterial effectiveness of the filters was evaluated following the Time-Kill Procedure (ASTM E2315), assessed by measuring the reduction of the microbial population over time. From visual inspection, the highest antimicrobial activity was observed for both Lot A and B photocatalytic filters. Results are reported in Table I. Figure 4 displays the results of the antibacterial tests carried out on the commercial filter and Lot A photocatalytic filter. It is worthy of note that already after 30 minutes of incubation, any viable bacteria was detected. The maximum efficiency was found already at the shortest duration and was maintained thereafter.

A time-killing effect was observed also for the commercial filter. Indeed, when in contact with this kind of filter, the number of viable cells decreased by up to 41% after 30 minutes of incubation, and it dropped to 36% after 60 minutes, before decreasing to very low levels only after 120 minutes.

VOC and TSP depolluting effectiveness

As regards the results of the experimentation of depolluting with the tested device, the results are shown in Table II and Figure 5 as regards the ability to decrease TSPs, and in Table III and Figure 6 with regard the ability to decrease VOC.

In Figure 5 it is apparent that, in the absence of ventilation (i.e., in static conditions), TSPs were roughly stable over the entire duration of the experiment, whereas a device equipped with Lot A photocatalytic filter induced a dramatic decrease in TSP load over time, until approaching 100% removal after 120 minutes.

Figure 6 shows the time course for VOCs. In the absence of ventilation (i.e., in static conditions), VOCs displayed a slow but significant decrease over time. In contrast, a ventilation system equipped with Lot A photocatalytic filter in the presence of UV irradiation (air purifier) induced an abrupt decrease in VOC content during the first 30 minutes, while

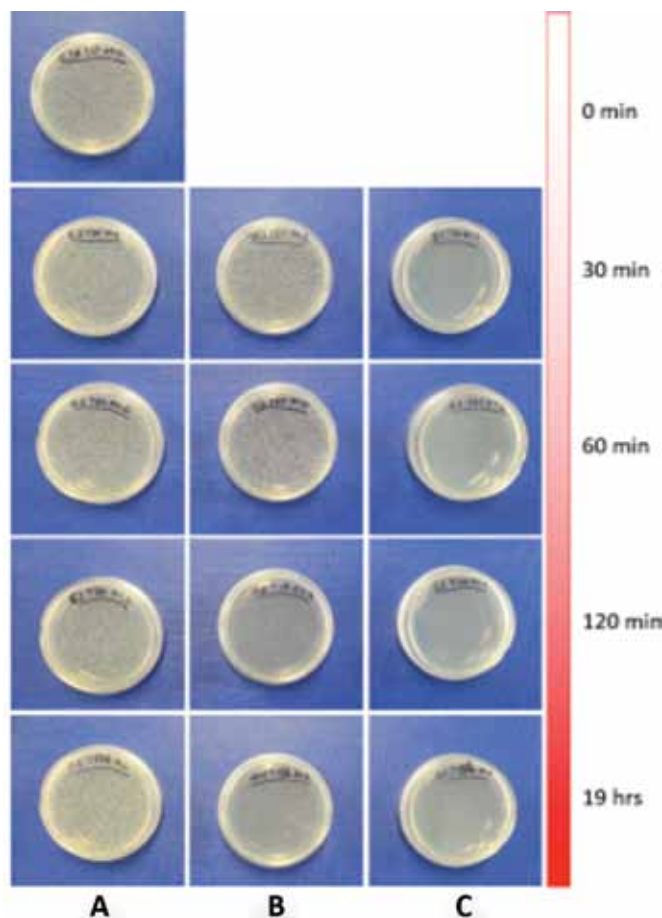


Fig. 4 - Quantification of colony forming units (CFUs) on Lysogeny broth agar (LBA) surfaces after keeping the bacteria in contact with different filters for variable durations: (A) control, (B) commercial filter and (C) Lot A photocatalytic filter.

the their content was roughly stable thereafter, likely because not all volatile chemicals had been photocatalytically oxidized.

Conclusions

Activated charcoal filters treated with TiO₂ and illuminated by UV LEDs to induce photocatalysis (supplied by NextMaterials) were found to be extremely effective in killing bacteria already after 30 minutes postexposure. In addition, they were found to be very effective against solid particulates, VOCs and bacteria, leading to their long-term inactivation. It is worthy of note that their abatement capacity was closely linked to the number of volume changes per hour, thus it was a function of the fan system used.

Overall, we have thus developed an air purifier prototype (92 × 92 × 25 mm³) consisting of 4 fan systems, activated charcoal filters treated with TiO₂ and UV LED lighting. The device is characterized by low noise and is able to reduce significantly the pollution caused by cigarette smoking (VOCs and TSP). By the same token, this device could be considered a suitable option to treat indoor environmental pollution (from vehicular traffic, cooking of food, etc.).

TABLE I - Effect of different sample filters on bacterial (*E. coli*) survival at different time intervals

	Sample							
	Control (%)		Commercial filter (%)		Lot A photocatalytic filter (%)		Lot B photocatalytic filter (%)	
0 min	100	±0	100	±0	100	±0	100	±0
30 min	100	±16	41	±10	0	±0	0	±0
60 min	100	±1	36	±11	0	±0	0	±0
120 min	100	±8	5	±1	0	±0	0	±0
19 hours	100	±20	0	±0	0	±0	0	±0

Results are expressed as means ± standard error of the mean (SEM). Control at any given time was set to 100%.



TABLE II - Results of the TSP reduction tests

	Sample			
	Static 1	Static 2	Air purifier 1	Air purifier 2
0 min	0	0	0	0
2 min	957	383	340	290
9 min	5,047	4,252	3,780	3,200
16 min	5,402	4,203	4,115	3,392
23 min	5,425	4,432	2,760	3,499
30 min	5,540	4,518	2,255	2,777
37 min	5,580	4,523	1,853	2,211
44 min	5,516	4,569	1,556	1,813
51 min	5,543	4,645	1,330	1,490
58 min	5,502	4,751	1,134	1,270
65 min	5,383	4,620	956	1,029
72 min	5,432	4,642	777	860
79 min	5,300	4,567	654	683
86 min	5,284	4,541	523	549
93 min	5,329	4,382	410	439
100 min	5,271	4,284	307	335
107 min	5,194	4,293	244	264
114 min	5,056	4,270	186	210
120 min	5,077	4,265	150	165

Results are expressed as $\mu\text{g}/\text{m}^3$ and were obtained from 2 independent measurements.

TSP = total solid particulates.

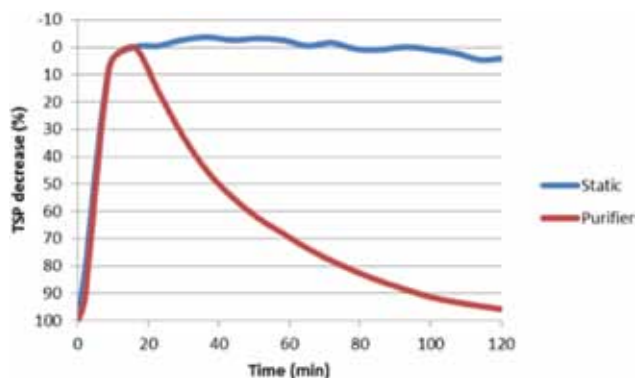


Fig. 5 - Results for total solid particulate (TSP) variations over time in static conditions and in the presence of the air purifier, which was a ventilation system equipped with Lot A photocatalytic filter in the presence of UV irradiation.

Disclosures

Financial support: No grants or funding have been received for this study.

Conflict of interest: None of the authors has any financial interest related to this study to disclose.

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TABLE III - Results of the VOC reduction tests

	Sample			
	Static 1	Static 2	Air purifier 1	Air purifier 2
0 min	7.61	3.39	2.15	3.12
2 min	15.66	10.93	9.81	10.23
9 min	39.43	34.62	28.43	30.01
16 min	39.70	36.54	32.74	32.18
23 min	39.13	36.50	29.90	30.64
30 min	38.46	36.10	23.55	27.43
37 min	37.92	35.08	22.17	26.53
44 min	37.40	34.91	21.56	26.08
51 min	36.92	34.53	21.23	25.83
58 min	36.47	34.07	21.09	25.64
65 min	35.95	33.47	20.97	25.50
72 min	35.67	33.13	20.87	25.40
79 min	35.30	32.75	20.75	25.31
86 min	34.99	32.43	20.74	25.27
93 min	34.70	31.89	20.69	25.20
100 min	34.37	31.61	20.62	25.12
107 min	34.16	31.37	20.59	25.08
114 min	33.92	31.14	20.55	25.03
120 min	33.71	30.95	20.54	25.00

Results are expressed as $\mu\text{g}/\text{m}^3$ and were obtained from 2 independent measurements.

VOCs = volatile organic compounds.

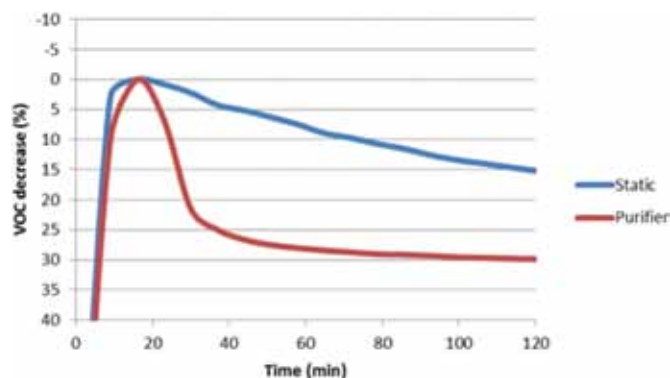


Fig. 6 - Results for volatile organic compound (VOC) variations over time in static conditions and in the presence of the air purifier, which was a ventilation system equipped with Lot A photocatalytic filter in the presence of UV irradiation.

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