Indoor PM2.5 removal efficiency of two different Non-Thermal Plasma systems

Hernández-Díaz, D.;[1*] Martos-Ferreira, D.;[2] Hernández-Abad, V.;[3] Villar-Ribera, R.;[4] Tarrés, Q.;[5] Rojas-Sola, J.I.[6]

1. Serra Húnter Programme, Department of Engineering Graphics and Design, Polytechnic University of Catalonia, TR5 Campus Terrassa, 08222 Terrassa, Spain; david.hernandez-diaz@upc.edu.
2. Department of Electronics, The Salesian University School of Sarrià, Passeig Sant Joan Bosco 74, 08017 Barcelona, Spain; d.martos.ferreira@outlook.com.
3. Department of Engineering Graphics and Design, Polytechnic University of Catalonia, TR5 Campus Terrassa, 08222 Terrassa, Spain; vicente.hernandez@upc.edu.
4. Department of Engineering Graphics and Design, Polytechnic University of Catalonia, Campus Manresa, 08242 Manresa, Spain; ricardo.villar@upc.edu.
5. LEPAMAP Group, Department of Chemical Engineering, University of Girona, 17003 Girona, Spain; joaquimagusti.tarres@udg.edu
6. Department of Engineering Graphics, Design and Projects, University of Jaén, 23071 Jaén, Spain; jirojas@ujaen.es.

[*] Correspondence: Polytechnic University of Catalonia, TR5 Campus Terrassa, 08222 Terrassa, Spain; david.hernandez-diaz@upc.edu; +34 937.398.662.

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Abstract

The use of non-thermal plasma (NTP) generators in air processing systems and their duct networks to improve indoor air quality (IAQ) has grown considerably in recent years. This paper reviews the advantages and disadvantages of NTP generators for IAQ improvement in biological, chemical and particulate pollutant terms. Also, it assesses and compares the ability of a multipin corona discharge (MPCD) and a dielectric barrier discharge (DBD) generator to reduce the concentration of fine particulate matter (PM2.5) in recycled, unfiltered air in a refrigeration chamber. The MPCD generator was found to have a higher PM2.5 removal efficiency; also, it was faster in removing pollutants, used less energy, and produced much less ozone. The fact that the MPCD generator performed better was seemingly the result of its increased ion production mainly. NTP generators, however, cannot match air filtration media purifiers in this respect as the latter are much more effective in removing particles. Besides, NTP-based air purifying technology continues to be subject to a major drawback, namely: the formation of ozone as a by-product. In any case, the ozone generation was uncorrelated to ion emission when using different technologies.

Keywords: Non-thermal plasma (NTP); Corona discharge; Dielectric barrier discharge (DBD); Fine particulate matter ($PM_{2.5}$); Ozone (O_3); Indoor Air Quality (IAQ).

1. Introduction

Indoor air quality (IAQ) is an issue of growing concern for researchers and legislators for various reasons especially prominent among which are the typically long exposure times of humans to polluted indoor air, the adverse effects of some air pollutants on health and well-being, and the high diversity and chemical complexity of air in enclosed spaces (Kelly and Fussell, 2019).

The use of atmospheric-pressure non-thermal plasma (NTP) generators in HVAC (heating, ventilation, and air-conditioning) systems, AHU (air handling units), purifiers and similar air processing devices, and also in their duct networks, has grown substantially over the last few years. NTP generators use an electric field to impart atoms and molecules in recycling air positive or negative charge (unipolar ionization), or both (bipolar ionization). The transfer of energy resulting from the collision of charged molecules with neutral molecules in air causes the formation of ions and free radicals. This process alters the natural ionic balance, which is estimated to be 1000 negative ions/cm^3 and 1200 positive ions/cm^3 (Laza and Lotrean, 2009). Also, the resulting ions and free radicals react with water molecules in air to form reactive oxygen species (ROS) including hydrogen peroxide (H_2O_2), ozone (O_3), hydroxyl radical (OH^-), superoxides (O_2^-) and singlet oxygen [(O_2(1Dg)] (Priya Arjunan and Morss Clyne, 2011). The range of ROS formed depends on the polarity of the reacting ions (Zhang et al., 2015), the design parameters of the NTP reactor used and the ambient conditions during the air ionization process.

The use of ROS to increase IAQ is grounded on the difficulty of the low- and medium-performance air purifying systems typically used for cost- and energy-efficiency reasons (Azimi and Stephens, 2013) to capture and remove biological pollutants. In fact, filters, condensate trays and ducts in such systems provide a fertile breeding ground for microbial reproduction and proliferation indoors. Although these systems have proved effective in eliminating a variety of microbes, the way they fulfil this goal is not fully understood (Zhou et al., 2018). The most widely accepted hypothesis is that they disrupt cells by causing the formation of hydroxyl radicals upon deposition of ions formed in the purification process onto the surface of membrane cells. Because hydroxyl radicals have a high affinity for hydrogen atoms, they react massively with them and irreversibly destroy membrane proteins and lipids to form water as a result (Digel et al., 2005). One alternative hypothesis holds that acquisition of electric charge by cell membranes to an extent exceeding their tensile strength causes their electrostatic disruption (Mendis et al., 2000). Still another hypothesis is that electrostatic interactions of the many charged groups depositing onto cell walls induce physiological changes in cells (Noyce and Hughes, 2002).

The ion flow produced by NTP generators has been shown to possess bactericidal properties against gram-positive and gram-negative bioaerosols settling on static physical media such as air filters and culture dishes, negative ions being less efficient than positive ions in this respect (Kim et al., 2016; Kim et al., 2011). Because they tend to produce ozone (O_3), which is a long-lived ROS with proven biocidal activity, negative ions were formerly assumed to be more efficient than positive ions. In systems generating ozone as a by-product, cell disruption has a weaker effect than the gas itself (Fletcher et al., 2007). When ozone production does not play the leading role in the process, the antibacterial efficiency of positive and negative ions increases with increasing ion concentration and exposure time (Kim et al., 2011).

Even more interesting than improved IAQ is antibacterial efficiency in single-step dynamic processes. For example, ions produced by a bipolar ionization NTP system have shown bactericidal action against *Staphylococcus epidermis*, their efficiency increasing with increasing polarity switching and decreasing with increasing air flow rate (Lee et al., 2014). As in static IAQ systems, the bactericidal efficiency of unipolar ion generators is seemingly greater with positive ions than it is with negative ions (Nunayon et al., 2019). In fact, the biocidal capacity of NTP generators producing negative ions has proved selective against some bacteria (Lai et al., 2016). All mathematical models used to simulate the behaviour of unipolar ions inside ducts with a view to predicting the zones of maximum biocidal action predict that antibacterial activity increases with decreasing air flow rate and distance from the ion emission source (Zhou et al., 2018; Zhou et al., 2016).

As far as antiviral action is concerned, bipolar ions have been reported to inactivate influenza virus H1N1 (by damaging hemagglutinin), and also to be effective against polio and Coxsackie viruses under laboratory conditions (Nishikawa and Nojima, 2003). Likewise, the ions produced by a carbon fibre-based corona discharge generator have proved effective against MS2 virus aerosolized on an air filter via an *Escherichia coli* host, their efficiency being greater with bipolar ionization than with unipolar ionization (Hyun et al., 2017). Moreover, single-step NTP systems have provided inactivation ratios for influenza virus H5N2 on a par with those of conventional UV light systems (Terrier et al., 2009). Thus, a DBD-based packed-bed NTP reactor with negative polarization proved highly efficient in inactivating MS2 phage, and porcine reproductive and respiratory syndrome virus (PRRSv), in aerosol. The degree of inactivation increased with increasing voltage and power, and peaked at 30 kV and 21 W, respectively, but was virtually independent of the air flow rate (Xia et al., 2019; Xia et al., 2020). These studies recently had some media coverage as a result of PRRSv belonging to the order *Nidovirales*, which includes severe acute respiratory syndrome coronavirus (SARS-CoV) (Jung et al., 2009).

In addition to biological pollutants, NTP systems are effective in removing or abating some organic volatile compounds (VOCs) in air. Using ion emitters at increased voltages and frequencies causes greater number of electrons to break molecular bonds in the pollutants, thereby increasing the efficiency with which they are removed (Byeon et al., 2010; Santos et al., 2020). On the other hand, the removal efficiency of NTP systems seemingly decreases with increasing initial concentration of VOCs and/or air flow rate (Fojlaley et al., 2014; Karatum and Deshusses, 2016; Vandenbroucke et al., 2011). By contrast, other VOC removal technologies such those based on adsorption onto activated carbon are less efficient with low VOC concentrations (Ragazzi et al., 2014; Schiavon et al., 2017). Because each type of molecular bond requires a minimum amount of energy to be broken (Urashima and Chang, 2000), the energy efficiency of

NTP systems is higher when the specific pollutant to be removed is known. The main problem with such systems in abating chemical pollutants arises from incomplete oxidation of gases and the formation of unwanted by-products (Vandenbroucke et al., 2011), which often include ozone (Guo et al., 2019; Holzer et al., 2018; Zhang and Jenkins, 2017). This gas can react with various VOCs, hydroxyl radical ($OH^·$) and nitrate radical ($NO_3^·$), in indoor air (Preis et al., 2013; Waring and Wells, 2015). The problem with the formation of ozone as a ionization by-product is not only its high reactivity against VOCs and free radicals, but also its potentially hazardous nature (EPA, 2013). As a result, ozone-producing IAQ improvement technologies can have adverse impacts on cardiovascular health (Day et al., 2018). In any case, removing VOCs with an NTP system does not require the presence of ozone (Kim et al., 2016); also, the main drawbacks of NTP technology for VOC removal can be minimized by using a catalyst filter made with manganese, titanium or doped titanium oxide (Chen et al., 2009). The combined technology is commonly known as Plasma-Assisted Catalysis (PAC) or Plasma-Driven Catalysis (PDC).

NTP systems, which are typically integrated in electrostatic precipitators (ESPs), have proved highly effective for capturing fine particulate matter ($PM_{2.5}$) in air — submicron particles included (Cho et al., 2012; Park et al., 2008). The greatest advantage of ESPs over other types of particle capture systems is that they use energy sparingly and have a small pressure drop (Mizuno, 2000). However, because ESP technology generally produces ozone and is expensive to maintain, it is scarcely used to purify indoor air (Huang et al., 2016; Shin et al., 2019). Particles in air are captured by electrostatic precipitation (Mizuno et al., 1999), the capturing efficiency increasing with increasing voltage (Byeon et al., 2006; Byeon et al., 2010; Thonglek and Kiatsiriroat, 2014). There is some scientific controversy over the particle removal efficiency and pulse rate of ESP systems. Thus, some studies indicate that increasing the frequency decreases the collection efficiency (Byeon et al., 2006; Byeon et al., 2010), whereas others suggest otherwise (Thonglek and Kiatsiriroat, 2014). The ability of ESP systems to remove fine and submicron particles is critically dependent of the air flow rate; this is so because maximizing collision between charged particles by increasing this variable can lead to an inadequate number of charged particles through a decreased residence time (Thonglek and Kiatsiriroat, 2014). This may also have been the source of the controversial results obtained as regards the influence of the pulse frequency.

NTP systems and conventional physical filters act synergistically. Thus, producing positive ions upstream filters has been found to increase their efficiency in direct proportion to the ion concentration and inverse proportion to the air flow rate (Park et al., 2011; Park et al., 2009). Likewise, negatively charging low-efficiency filters has been shown to increase their capacity to capture bacteria and fungal spores (Huang et al., 2008). There is scientific evidence that NTP systems for IAQ improvement are effective for removing microbiological pollutants, VOCs and PM. Such evidence, however, was obtained under strictly controlled laboratory conditions. In theory, bipolar ionization NTP systems should be able to capture fine particulate matter ($PM_{2.5}$) without the need for electrostatic collectors or physical filters (viz., simply by agglomeration and subsequent natural precipitation of particles) (Thonglek and Kiatsiriroat, 2014). Whether a single use of NTP technology suffices to remove suspended particles was questioned in a recent review (Liu et al., 2017).

Because indoor air quality (IAQ) affects public health and no existing technology is effective enough to completely purify air in any type of environment, it is important to further investigate those specific technologies with the highest potential in order to identify the particular scenarios where they can provide the highest benefit–cost ratio. Bipolar ionization NTP technology has proved a promising choice for removing various types of pollutants that detract from IAQ. Although it is subject to some shortcomings, it has proved unequivocally effective under laboratory conditions. This has promoted the incorporation of various forms of NTP devices into ventilation, air-conditioning, air purification systems and, especially, air duct networks, frequently on purely commercial grounds (i.e., without scientific support for their removal efficiency or their actual benefits in relation to their shortcomings). Not for nothing has the European Union established restrictions of the production of radicals for biocidal purposes (European Union, 2012). Also, a recent investigation by a panel of experts found that no conclusive studies on the ability of NTP technology to improve air quality as assessed in chamber or decay tests were available (Zhang et al., 2011). A gap of knowledge therefore exists that prevents establishing reliable application patterns for NTP technology under real-life conditions —a purpose for which the single-pass test falls short— in order to facilitate its efficient, safe use in ventilation, air-conditioning and air purification systems.

The primary purpose of this work was to assess the efficiency in continuous operation of two bipolar ionization systems based on multipin corona discharge (MPCD) and dielectric barrier discharge (DBD) in reducing the concentration of PM2.5 in indoor air in a semi-real environment. Comparing these two NTP technologies is in itself a major novelty because, to the authors' knowledge, they have never to date been contrasted under semi-real conditions despite being the most extensively researched in connection with air purification. Also, continuously monitoring their operation is expected to provide a better understanding of their mechanism of action with a view to allowing future research to be aimed at specific applications where NTP technology can perform better to improve IAQ. The choice of $PM_{2.5}$ as the target for removal was justified by its being the most robust indicator of long-term mortality from exposure to polluted air (Chan et al., 2015). In fact, $PM_{2.5}$ has been associated to increased mortality and morbidity of chronic pulmonary diseases (Chi et al., 2019), deterioration of lung function in pregnant women (Mulenga and Siziya, 2019), and several millions of premature deaths each year (I. Straif et al., 2013). This has led to $PM_{2.5}$ being globally used as an indicator for external and internal air quality monitoring, even in the form of bio-indicators such as Hybrid Petunia (Benaissa et al., 2019). One other purpose was to monitor the concentration of ozone —production of which is the main shortcoming of NTP systems— during the process.

The remainder of the paper is structured as follows: Section 2 describes the methodology and specifies the materials used; Section 3 reflects on the results of the experiment; and Section 4 draws interesting conclusions from the results.

2. Materials and methods

The experiment was conducted in a 6 m^3 (2 m × 2 m × 1.5 m) refrigeration chamber located in an air-conditioned room where the temperature and relative humidity were kept at (22 ± 2) °C and (50 ± 5) %, respectively, throughout. Placing the chamber in an air-conditioned room rather than conditioning the chamber itself was intended to avoid introducing an additional air flow that might have perturbed mixing of the resulting particles.

The experiment involved a total of 7 tests. The first three were control tests where no NTP generator was in operation. A bipolar ionization multipin-based corona discharge NTP generator (viz., an AP-DC5601 model from Shanghai Anping Static Technology Co. Ltd., Shanghai, China) was used in two other tests, and a bipolar ionization dielectric barrier discharge (DBD) NTP generator (viz., a BXMCB2 model from Bioxigen S.r.l., Cologna Veneta, Italy) in the remaining two. Table 1 shows the experimental units in operation in each test. The system included a particle generation unit, a device for mixing and spreading particles in the chamber inner air, an NTP generator —in the tests where one was used— and sensors to monitor the concentrations of $PM_{2.5}$ and ozone in air.

Table 1. Units in operation in each test.

Test	Smoke generator	Fan	System A	System B	Sensors
1	ON	ON	OFF	OFF	ON
2	ON	ON	OFF	OFF	ON
3	ON	ON	OFF	OFF	ON
4	ON	ON	ON	OFF	ON
5	ON	ON	ON	OFF	ON
6	ON	ON	OFF	ON	ON
7	ON	ON	OFF	ON	ON

Particles were produced by using Nagchampa incense wicks from Goloka Seva Trust (Bangalore, India), which are a common choice for regular incense users. Particles were mixed and spread by using a MU-TT 200 axial fan from Salvador Escoda, S.A. (Barcelona, Spain), operating at a flow rate of 830 m^3/h. The ozone concentration was measured in a continuous manner by using an Advanced Sense IAQ probe from GrayWolf Sensing Solutions, Ltd. (Annacotty, Ireland), whereas the mass concentration of suspended particles was measured at 12-min intervals by using a DSM 501 sensor from Samyoung S & C (Gyeongii-do, Korea). Because the DSM 501 sensor is based on light scattering, not gravimetric analysis, particle mass concentrations were estimated from Eq. (1) (Franken et al., 2019; Hinds, 1982).

$$C_m = 10^{-15} \cdot \rho_p \cdot C_n \cdot \frac{\pi}{6} \cdot d^3{}_{m/a} \tag{1}$$

Where C_m is the $PM_{2.5}$ mass concentration (µg/m3), C_n the particle number concentration (pcs/cm3), ρ_p the estimated incense smoke density (1.1 g/cm3), and dm/a the mean diameter of incense smoke particles in the range of $PM_{2.5}$, which was obtained from reported data (Ji et al., 2010).

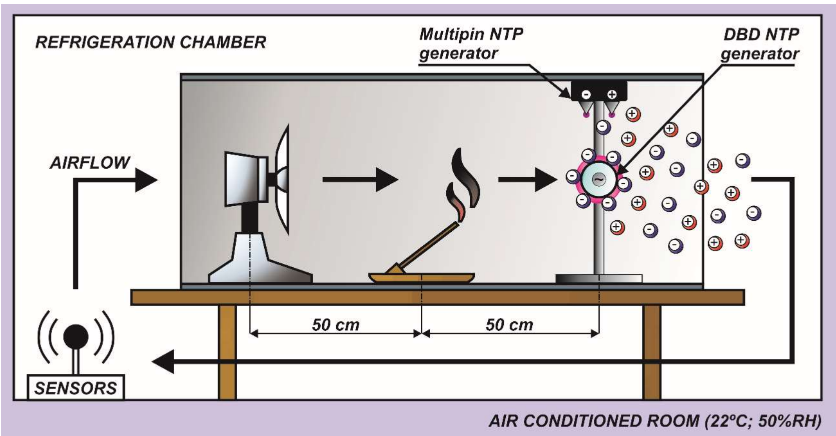

Fig. 1 Experimental set-up

Figure 1 depicts the different elements of the experimental set-up. The fan, wicks and plasma generators were placed inside a 120 cm long × 50 cm wide × 50 cm high galvanized metal duct on an 80 cm high table, whereas the sensors were positioned on the ground in a corner of the refrigeration chamber.

The plasma generators were placed according to the manufacturers' instructions (viz., normal to the direction of the fan air flow). The MPCD generator was positioned in such a way that most of the air flow circulated beneath the ion emitters, whereas the DBD generator was placed halfway between the floor and ceiling of the duct so that half the air flow would circulate beneath and half above it.

Each test included monitoring the $PM_{2.5}$ and ozone concentrations for 8 h. This was followed by evacuation of polluted air from the chamber to renew its atmosphere and by nebulization with 70% ethanol for 1 h to clean all inner surfaces. Experimental configurations were set up, and the temperature, relative humidity and $PM_{2.5}$ concentration of the chamber and room were allowed to equilibrate prior to each monitoring event. Also, monitoring measurements were made after the fan, plasma generator —if one was used— and sensing system were started; then, the wicks were ignited and the door was closed until the process was finished and all units were switched off. Plasma generators, when used, worked throughout the test. The procedure is depicted in Fig. 2.

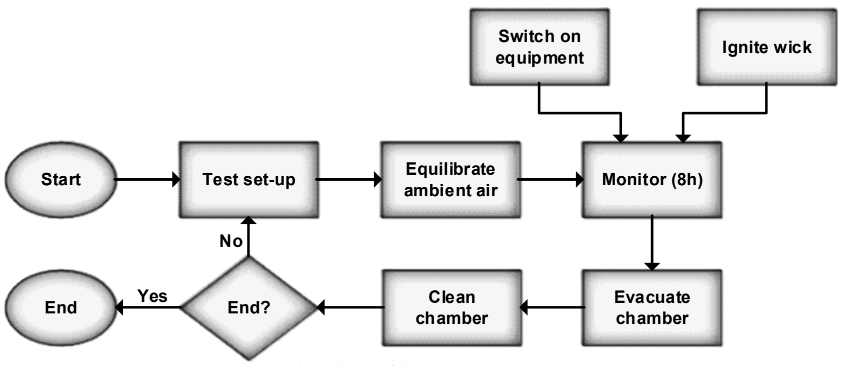

Fig. 2. Flow chart of the experimental procedure.

2.1 Bipolar multipin corona discharge NTP generator (MPCD generator)

The operating principle of this system is bipolar ionization of air by effect of corona discharges from many pins (electrodes). The generator used a power supply (220 V AC, 50 Hz, 5 W) and a 50 cm long electrostatic bar consisting largely of aluminium profile.

The profile was encapsulated in plastic and used to hold the electrical components. Each end of the bar was fitted with a plastic cap that was screwed to the profile. The bar had a positive and a negative ion emitter powered by a DC power supply, and the emitters were uniformly distributed steel tips. Both types of emitters were arranged in a staggered pattern (Fig. 3).

The output voltage at the emitter tips was measured with an 80K-40 high-voltage probe from Fluke Corp. (Everett, WA, USA) connected to a DSOX3054T oscilloscope from Keysight Technologies, Inc. (Santa Rosa, CA, USA), and found to be +8 kV in the positive emission band and –8 kV in the negative band. The concentration of positive and negative ions at a distance of 200 mm from the generator as measured with an AIC20M ion counter from Alphalab, Inc. (Salt Lake City, UT, USA) with 25% precision was 1.9×10^7 and 9.3×10^7 ions/cm^3, respectively.

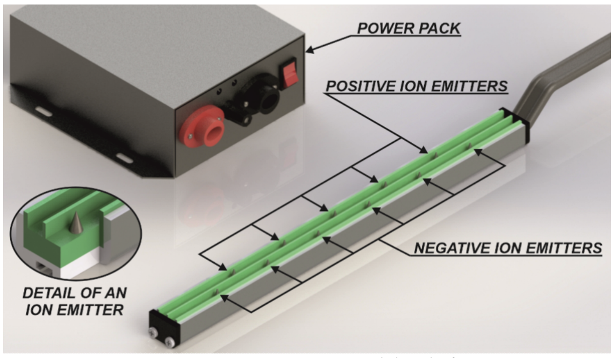

Fig. 3. MPCD generator. Components and detail of an ion emitter.

2.2. Bipolar dielectric barrier discharge NTP generator (DBD generator)

This generator (Fig. 4) consists of two jointly assembled modules, namely: an ion emitter and a power supply (230 V AC, 50 Hz, 6 W). The two can be readily disassembled for maintenance or replacement. The dielectric is a glass cylinder 180 mm long × 40 mm wide with spherical ends. An also cylindrical piece of metal about 10 mm wide that is concentrically inserted in the tube and connected to the power supply acts as inner electrode. Most of the outer surface of the tube is covered by a metal mesh acting as ground electrode.

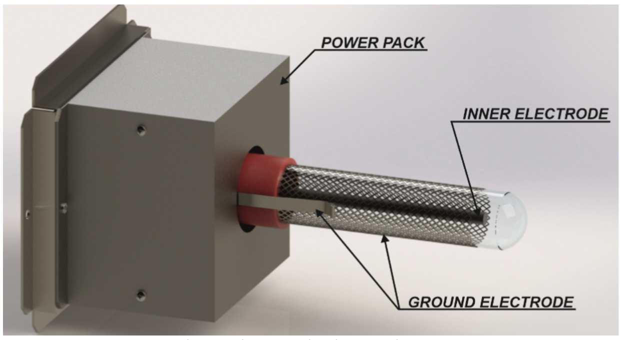

Fig. 4. Dielectric barrier discharge plasma generator.

The output voltage of the emitter could not be measured because this component was inaccessible with the system in operation. According to the manufacturer, the system uses pulsating AC. The concentrations of negative and positive ions were ~15% and ~35% lower, respectively, than those obtained with the CD generator under the same conditions.

Silent and corona-based discharge are the only two types of discharge capable of producing non-thermal plasma under atmospheric pressure and ambient temperature conditions (Yehia and Mizuno, 2013). A multipin generator ionizes air through a corona discharge, whereas a DBD generator with a coaxial cylinder and the dielectric in contact with the inner side of the outer electrode can ionize air both by corona discharge and by silent discharge depending on the ratio between radii (Yehia, 2008). The DBD generator used in this work ionized air by corona discharge (CD) only and was hence based on the same physical principle as the other. Also, the MPCD generator used emitter tips directly wired to the power supply to produce positive and negative ions in a continuous manner. On the other hand, the DBD-based generator used a cylindrical metal mesh as a part of an electrode arrangement including insulating material in the discharge pathway (Brandenburg, 2017). The latter type of generator operated on pulsed voltage.

3. Results and discussion

The experimental set-up was used to compare the efficiency of the two types of NTP generators in reducing atmospheric $PM_{2.5}$ levels in a closed room with 100% air recycling but no physical filters or electrostatic collectors. Additional measurements included ozone production as an unwanted outcome.

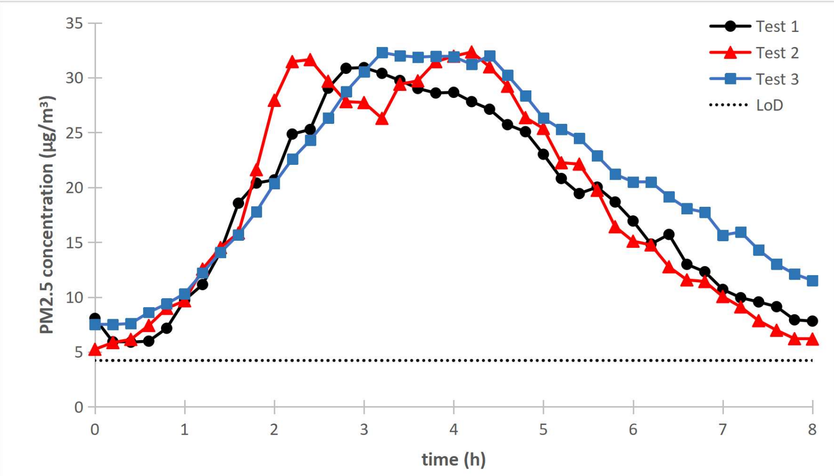

Fig. 5. Temporal variation of the $PM_{2.5}$ concentration in the chamber during the tests without generators.

Three tests were conducted with no generator to check the goodness of the experimental procedure and provide reference values for comparison with the two generators. Figure 5 shows the variation of the $PM_{2.5}$ mass concentration inside the refrigeration chamber in such tests. The three curves were rather similar and hence suggestive of a high repeatability in the experimental procedure. As can be seen, the $PM_{2.5}$ concentration initially increased as the obvious result of combustion of the wicks, then levelled off at near-final values and eventually fell to near-initial levels. According to the manufacturer, each wick should have provided 30–45 min of smoke; however, the $PM_{2.5}$ concentration increased over a period of 2–3 h. Because it is relevant to the conclusions of this study, the figures also show the limit of detection (LOD) of the sensor. Although the reported LOD (4.28 μg/m^3) is fairly low for an inexpensive sensor, the features of the sensor do not make it suitable for measurements in very clean air environments (Wang et al., 2015).

Fig. 6 shows the variation of the $PM_{2.5}$ mass concentration inside the chamber in the presence of the MPCD generator and compares it with the results of the previous (control) tests with no generator. As can be seen, the highest concentration found with the generator in operation was 39.2 and 35.6% lower than that obtained in the control tests. However, if only the amounts of pollutant produced by the incense wicks are considered, the peaks of the curves for Tests 4 and 5 were 47 and 49.1% smaller, respectively, than those of the control curve. Also, based on WHO's recommendations, which include a maximum daily average of 25 $\mu g/m^3$ and an annual average of 10 $\mu g/m^3$ for $PM_{2.5}$ (Organization, 2006), the former and latter limit were exceeded 35% and 77.5%, respectively, of the time in the control tests. On the other hand, the 25 $\mu g/m^3$ limit was never exceeded with the MPCD generator and the 10 $\mu g/m^3$ limit was only surpassed 31.25% of the time on average by the two tests in combination. One other interesting result was the variation of $PM_{2.5}$ beyond the peak in each test. Thus, the average residence time for the control tests was close to the peak value. On the other hand, $PM_{2.5}$ started to fall immediately upon peaking with the MPCD generator. Therefore, the generator decreased the $PM_{2.5}$ concentration peak and accelerated recovery of the initial concentration as compared to the natural decay reflected in the control curves.

The $PM_{2.5}$ minima observed in Tests 4 and 5 were slightly lower than the initial pollutant concentrations. Because of the characteristics of the sensor, and of the similarity of such concentrations to the LOD, it was impossible to know whether the corona-discharge generator could have continued to reduce the $PM_{2.5}$ concentration in air below the initial values. In any case, the final values obtained with that generator in operation, and those provided by the DBD reactor, were typical of a standard clean environment.

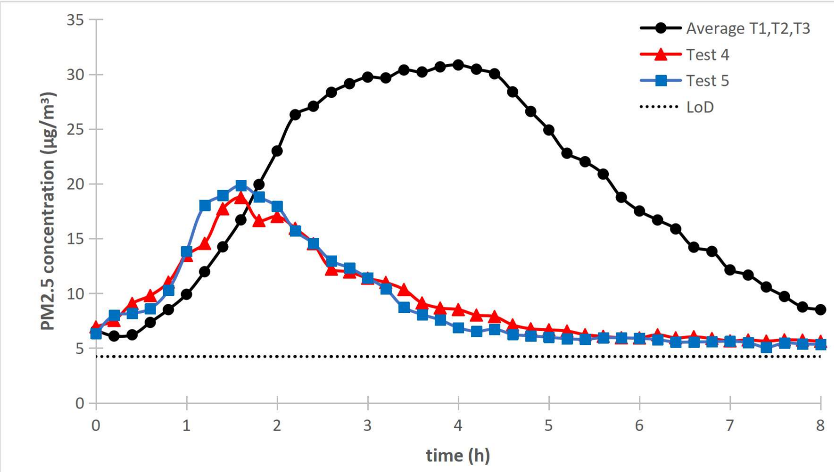

Fig. 6. Comparison of the temporal variation of the $PM_{2.5}$ concentration in the chamber with the corona discharge generator in operation and the average results of the three control tests (no generator).

Fig. 7 shows the variation of the $PM_{2.5}$ mass concentration in the refrigeration chamber in the two tests performed with the dielectric barrier discharge (DBD) plasma generator as compared with the results of the control tests. Again, the $PM_{2.5}$ concentration peaks were markedly smaller with the generator in operation, which reduced $PM_{2.5}$ levels by 37.7 and 47.4%, respectively in the two tests. Excluding the initial concentration of particles in the refrigeration chamber—the particles were not produced by the incense wicks— reduced the peak $PM_{2.5}$ concentrations in Tests 6 and 7 by 47.2 and 56.5%, respectively. Although the $PM_{2.5}$ concentration was reduced to a similar extent by both types of systems, the peak concentration was reduced 3.8% more by the DBD generator than by the MPCD generator. Based on WHO's reference levels, the DBD-based NTP generator ensured that the maximum recommended daily level (25 $\mu g/m^3$) was never exceeded. However, the annual limit (10 $\mu g/m^3$) was exceeded 46.25% of the testing time on average (i.e., the $PM_{2.5}$ concentration was above the daily limit 1.2 h longer with the DBD generator than it was with the corona-discharge generator). As can be seen from Fig. 7, the decrease in $PM_{2.5}$ achieved with the DBD-based generator was progressive and the $PM_{2.5}$ concentration levels remained near the peak value for a longer time than with the MPCD generator.

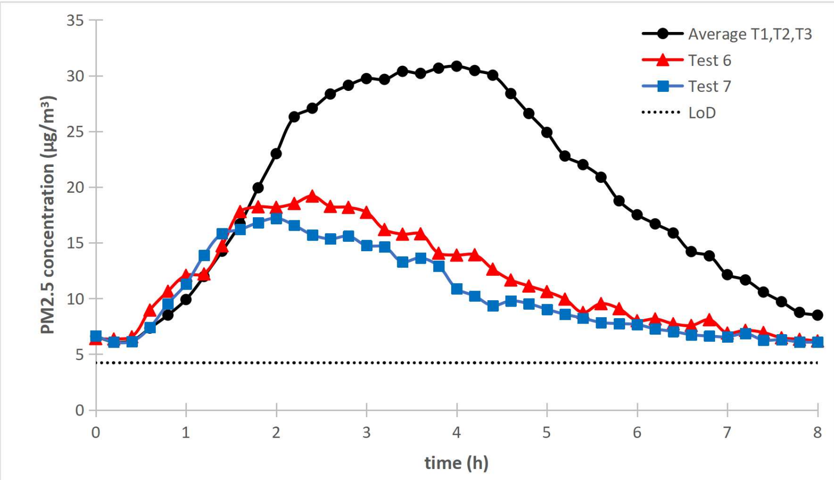

Fig. 7. Comparison of the temporal variation of the $PM_{2.5}$ concentration in the chamber with the dielectric barrier discharge generator in operation and the average results of the three control tests (no generator).

Fig. 8 shows the temporal variation of the $PM_{2.5}$ average abatement efficiency of the MPCD and DBD generators relative to natural decay. The removal efficiency was calculated from Eq. (2).

$$\eta_{PM2.5} = 1 - \frac{C\,(after\ plasma\ exposure)}{C\,(natual\ decay)} \quad (2)$$

Where the numerator represents the $PM_{2.5}$ concentrations due to wick combustion measured in the tests with the NTPs in operation and the denominator the average concentration induced by the wicks in the absence of the NTP reactors.

As can be seen, both types of generator were very similarly efficient in removing the pollutant during the initial stage of combustion and rise of the $PM_{2.5}$ concentration in the refrigeration chamber (viz., during the first 2 h of each test). Specifically, the $PM_{2.5}$ concentration at the end of this stage was approximately 33% lower than in the absence of the NTP generators. Beyond that point (viz., during the third hour of operation), the MPCD generator was 21% more efficient than the DBD generator. This difference persisted until all $PM_{2.5}$ produced by the burning of incense was removed —which took

4.5 h with the CD reactor. With no NTP generator, the average $PM_{2.5}$ concentration after 4.5 h was only 8% lower than the peak value. The DBD generator took 7 h to completely remove the amount of pollutant produced by the incense wicks. Although the average curve for the control tests did not reach the initial $PM_{2.5}$ level after 8 h, such a level was restored exactly after 8 h in Test 2. Also, although the DBD generator proved less efficient in this respect, it reduced the initial $PM_{2.5}$ concentration by 80% within 4.5 h.

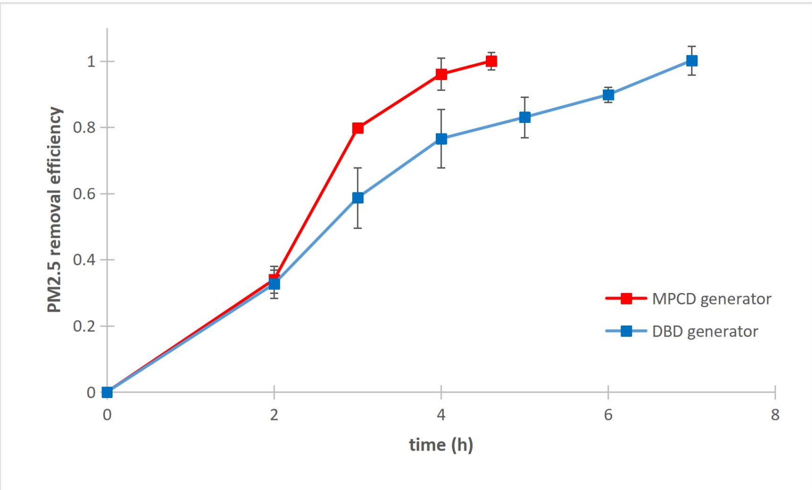

Fig. 8. Average efficiency of the corona and dielectric barrier discharge generators in reducing the $PM_{2.5}$ concentration generated by wicks.

In summary, both types of NTP generator reduced the peak level of particles by about 50% during the incense burning and pollutant spreading stage. Then, the MPCD generator reduced pollutant levels to a greater extent than the DBD reactor and completely removed the pollutant within 2.4 h on average. This was the likely result of the MPCD generator producing greater amounts of positive and negative ions. A similar study on unipolar technology suggested that the ion concentration could play a key role in the particle reduction efficiency of a system (Grinshpun et al., 2005). As the DBD generator was more energy-demanding, it was clearly less efficient in removing $PM_{2.5}$ from incense burning. DBD generators operating through a corona discharge (CD) are in fact known to be scarcely efficient for this purpose, as a result of the volume of plasma produced over the surface of the electrodes is lower than that confined between them (Yehia, 2008, 2019). In any case, the results suggest that this multipin technology provides greater benefits than that based on a coaxial cylinder dielectric.

As can also be inferred from the results, the difference in efficiency between the two types of generator decreased with increasing pollutant concentration. This suggests saturation at some point and also that, in the absence of filters, NTP generators must be scarcely efficient in removing particles from clean environments containing few ions. In fact, the large average distance between particles at low pollutant concentration must have hindered adhesion and subsequent precipitation. In any case, this conclusion is unwarranted owing to the low LOD of the sensor used.

Directly comparing our results with those of other studies or systems is hindered by the absence of conclusive efficiency data for NTP technology as assessed with the chamber test (Zhang et al., 2011) and also by differences among experimental set-ups. As shown here, however, both NTP systems had a very low Clean Air Delivery Rate (CADR) and One-Pass Single Efficiency (OPSE); in fact, both needed a large number of air recirculation cycles to completely remove the pollutant (see Table 2). Therefore, the particle removal efficiency of NTP technology is not comparable to its filtering efficiency as filtering can dramatically reduce PM levels (Du et al., 2011).

Table 2. Clean Air Delivery Rate and One-Pass Single Efficiency of each type of NTP generator.

NTP Generator	K_t (min^{-1})	K_n (min^{-1})	CADR (m^3/min)	OPSE (%)
MPCD	6.3×10^{-3}	1.1×10^{-3}	3.12×10^{-2}	0.23
DBD	2.2×10^{-3}	1.1×10^{-3}	0.66×10^{-2}	0.05

K_t decay constant, K_n natural decay rate constant. CADR and OPSE were calculated from ANSI/AHAM AC-1-2006 standard equations.

In regard to ozone production, Fig. 9 compares the average variation of the O_3 concentration in the two tests with the MPCD generator and those with the DBD generator. As can be seen, the DBD generator produced much greater amounts of ozone than did the MPCD generator, which indicates that ozone generation was uncorrelated with ion emission when using different technologies. Specifically, the ozone production ratio was 1.0 mg/h with the MPCD generator and 8.6 mg/h with the DBD generator. Although both ratios are low, they are high enough for the ozone concentration in a small room to exceed WHO's maximum allowable exposure limit: 5×10^{-2} ppm (0.1 mg/m^3) (Organization, 2006). In fact, the O_3 concentrations measured with the DBD generator in operation exceeded 5 ppm, a definitely hazardous level (Salonen et al., 2018). The MPCD generator produced less ozone—and probably would have produced virtually none if the metal ion emitters used had been replaced with carbon fibre emitters (Han et al., 2009; Han et al., 2008; Shin et al., 2019). Also, altering some design parameters of the DBD generator such as the length of the inner electrode, configuration of the ground electrode or overall reactor size might have substantially reduced its ozone production levels (Bahri et al., 2016). The joint use of a DBD generator and a UV photocatalytic system was previously found to increase the efficiency with which some organic and inorganic pollutants can be removed from air, and also to boost the decomposition of ozone through the following reaction (Abou Saoud et al., 2017; Abou Saoud et al., 2018; Assadi et al., 2014; Maciuca et al., 2012; Taranto et al., 2007):

$$O_3 + hv\,(254 < \lambda < 350nm) \rightarrow O_2 + {}^{\cdot}O$$

Catalytic oxidation with manganese oxide (MnO) or manganese doped with cerium oxide ($MnCeO_x$) recently proved highly efficient in decomposing ozone and other major pollutants in indoor air (Zhang et al., 2019). In those indoor air uses where air at a temperature higher than 90 °C is available (e.g., in airplanes, where fresh air comes from the lower stage of the engine), a palladium catalyst supported on activated carbon fibre (Pd/ACFs) can substantially improve ozone removal efficiency (Wu et al., 2017). Catalysts based on transition metal oxides are less efficient in this respect than are those using noble metals; however, the former tend to be preferred owing to their lower production costs (Yu et al., 2019). In any case, we must insist that the success of NTP technology in improving IAQ rests on completely avoiding ozone production, whether voluntarily or involuntarily. Otherwise, NTP technology will be subject to the same problems as conventional ESP systems, and also to those arising from the use of catalysts (e.g., pressure drops).

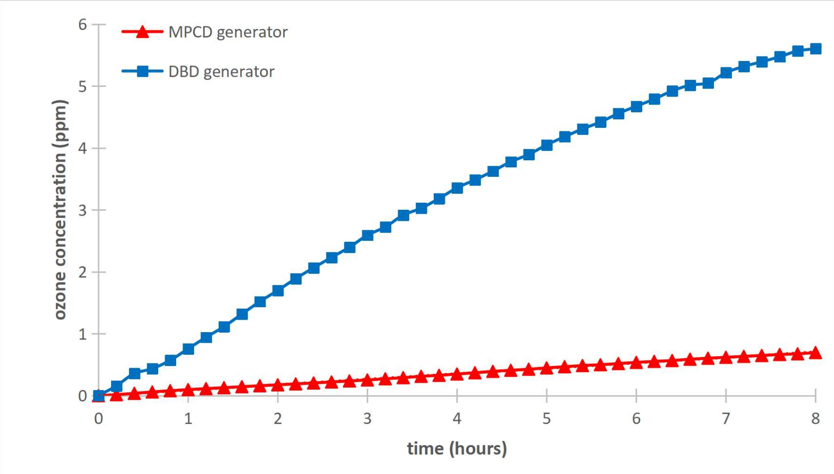

Fig. 9. Average variation of the ozone concentration in the chamber with the two types of generator in operation.

4. Conclusions

Atmospheric-pressure NTP generators have proved effective to remove various types of pollutants from indoor air, especially under controlled laboratory conditions. However, they can cause the formation of ozone as an unwanted ionization by-product.

The two types of NTP generators studied here (viz., a multipin corona discharge and a dielectric barrier discharge generator) succeeded in reducing the mass concentration of $PM_{2.5}$ in nearly completely recycled air with the need for no physical filter or electrostatic collector. Compared to other air purification choices such as physical filtration media or electrostatic collectors, NTP systems by themselves are rather inefficient in reducing $PM_{2.5}$ mass concentrations. However, NTP technology can considerably dampen pollutant concentration peaks and to reduce the time that particles are suspended in the air before starting to decay, which could be useful for very specific purposes or conditions.

Although both types of generators reduced $PM_{2.5}$ peak levels to a similar extent, the MPCD generator was clearly more efficient, energy-saving and faster than the DBD generator; also, the former produced less ozone. The experimental results suggest the presence of an ion saturation point at a given $PM_{2.5}$ concentration and air volume where more ions make no difference. At low particle concentrations, however, the efficiency of the system is clearly dependent on the production of an increased number of ions. This results, however, requires checking because, should it be confirmed, it would make small household bipolar generators useless for particle removal since, unless they are used in combination with filters or in places with high particulate matter loads, they will produce much smaller amounts of ions than the two types of generators studied here. Besides being useless for air particle treatment, they may be unsafe owing to the potential production of harmful by-products. The technological limitations of the sensor used in the research also make it advisable to repeat the tests with a highly sensitive PM2.5 and lower LOD.

In our humble opinion, NTP systems should not be used for particulate capture or removal unless in conjunction with conventional filtration devices to increase the collection efficiency of physical filtration or reduce pressure drops without detracting from collection efficiency. In very specific situations (e.g., when using physical filters is discouraged or where polluting particles are present only intermittently), NTP technology can be used by itself to improve air quality provided the typical concentration of pollutant and the effective dose of ions are accurately known.

NTP generators produce ozone and thus call for effective technical measures to avoid exceeding the legally tolerated levels of this gas and to prevent damage or discomfort to building occupants. If NTP generators are to be widely implemented as IAQ improvers, whether alone or in combination with other technologies, they should first be refined and guaranteed to produce no ozone.

Author contributions

David Hernández-Díaz wrote the manuscript and analysed the results; David Martos-Ferreira calibrated sensors and built the experimental set-up. Ricardo Villar-Ribera; Vicente Hernández-Abad made the figures; Quim Tarrés helped with the chemical comprehension and sustainability of the technologies; and José Ignacio Rojas-Sola supervised the study.

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Conflicts of Interest

The authors declare no conflict of interests.

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