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RESEARCH ARTICLE

OZONE GENERATION BY VACUUM ULTRAVIOLET RADIATION (λ = 172 NM) IN A CONTEXT OF DECENTRALIZED DRINKING WATER PRODUCTION

GENERACIÓN DE OZONO POR RADIACIÓN ULTRAVIOLETA AL VACÍO (λ = 172 NM) EN UN CONTEXTO DE PRODUCCIÓN DESCENTRALIZADA DE AGUA POTABLE

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ABSTRACT

The main problem with centralized drinking water production in isolated communities is the recontamination of water during distribution and storage. Therefore, a disinfection step at the point of consumption is important. A disinfection module, on a household scale, with a vacuum ultraviolet ozone generator operating with ambient air and powered by a 24 V source was built. The aim was to evaluate the production capacity of the photochemical ozone generator fed with ambient air without a gas dryer. The effect of relative humidity (RH) in air on photochemical ozone generation at different gas flow rates was investigated. The maximum ozone outputs with extra-dry air were 2.8 g-O_3/h and 3.7 g-O_3/h at lamp power levels of 20 W and 35 W, respectively (specific energy consumption: 7.1 kWh/kg-O_3 and 9.4 kWh/kg-O_3). The generator fed with water-saturated air produces nitric acid-free gas with ozone productions of 0.15 g-O_3/h to 0.79 g-O_3/h (25.3 kWh/kg-O_3). As the gas flow rate increases, ozone production rises until a plateau is reached. Increasing of the RH of the gas from 0 % to 20 % reduces ozone production by 35 % to 57 % for the lamp set at 20 W. A similar reduction occurred for 35 W. As the RH rises from 20 % to 100 %, the ozone output of the 20 and 35 W lamp plateaus at a non-zero value for each gas flow rate. The absence of nitric acid in the gas produced from humid air validates the use of the photochemical ozone generator in a decentralized drinking water production system.

Keywords: Decentralized water production; ozone; vacuum ultraviolet; water vapor.

RESUMEN

El principal problema de la producción centralizada de agua potable en comunidades aisladas es la recontaminación del agua durante su distribución y almacenamiento. Por lo tanto, es importante la desinfección en el punto de consumo. Se construyó un módulo de desinfección a escala doméstica con un generador de ozono ultravioleta al vacío que funciona con aire ambiente y se alimenta a una fuente de 24 V. El objetivo fue evaluar la capacidad de producción del generador de ozono fotoquímico alimentado con aire ambiente sin secador de gas. Se investigó el efecto de la humedad relativa (HR) del aire en la generación de ozono fotoquímico a diferentes caudales de gas. Las emisiones máximas de ozono con aire extraseco fueron de 2,8 g-O_3/h y 3,7 g-O_3/h con potencias de lámpara de 20 W y 35 W, respectivamente (consumo específico de energía: 7,1 kWh/kg-O_3 y 9,4 kWh/kg-O_3). El generador alimentado con aire saturado con agua produce gas sin ácido nítrico con producciones de ozono de 0,15 g-O_3/h a 0,79 g-O_3/h (25,3 kWh/kg-O_3). A medida que aumenta el caudal de gas, la producción de ozono se eleva hasta alcanzar una meseta. El aumento de la HR del gas del 0 % al 20 % reduce la producción de ozono entre un 35 % y un 57 % con la lámpara de 20 W. Se observó una reducción similar con la de 35 W. Al aumentar la HR del 20 % al 100 %, la producción de ozono de las lámparas de 20 y 35 W se estabiliza en un valor distinto de cero para cada caudal de gas. La ausencia de ácido nítrico en el gas producido a partir de aire húmedo valida el uso del generador de ozono fotoquímico en un sistema descentralizado de producción de agua potable.

Palabras claves: Producción descentralizada de agua; ozono; ultravioleta al vacío; vapor de agua.

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INTRODUCTION

Conventional drinking water production systems are centralized and rely on massive water treatment and distribution infrastructures that serve dense populations (Leigh & Lee, 2019; Rabaey et al., 2020). These systems follow a linear approach where water is treated near the point of abstraction and distributed in pipes or with water trucks over long distances before being used by residents (Chen, Wu, et al., 2017; Lakho et al., 2022). Contamination of water in a centralized drinking water production system during distribution as well as during storage is a significant problem (Balasooriya et al., 2023; Rusca et al., 2022; Wright et al., 2018). When treated water is transported over long distances and stored for extended periods, it is at risk of contamination by pathogenic microorganisms and by pesticides (Balasooriya et al., 2023; Rusca et al., 2022; Sarkar et al., 2022). This post-treatment recontamination highlights the importance of integrating a disinfection step directly at the point of consumption, i.e. a decentralized disinfection process on a household scale. Decentralized systems bring the treatment process closer to the point of consumption (Garrido-Baserba et al., 2022; Hafeez et al., 2021). However, the decentralization of disinfection processes is complexified by factors that are specific to rural and isolated communities. These include difficulties in sourcing chemical products, unstable energy distribution networks and low population density (Lei et al., 2021; Von Gunten, 2018). Ozone disinfection has previously been applied in decentralized settings (Dorevitch et al., 2020; Hendrickson et al., 2020; Lei et al., 2021).

Conventional dielectric barrier discharge (DBD) ozone generators require a high-voltage power supply and a system that produces dry carrier gas (air or pure oxygen). Microplasma ozone generators function at lower voltage, but still rely on electrical discharges to generate ozone (Dorevitch et al., 2020; Hendrickson et al., 2020). The presence of water in the carrier gas promotes the generation of nitric acid (HNO_3) in discharge ozone generators and damages the devices (Gottschalk et al., 2010; Rakness, 2015). Indeed, the electrical discharges dissociate molecular nitrogen (N_2) to produce NO_x which then react with water molecules to produce HNO_3. In addition, high concentration of nitrates (NO_3^-) in drinking water poses a danger to human health (Health Canada, 2013; World Health Organization, 2022). Air drying has an estimated energy consumption of around 5.5 W/m^3 to 10.4 W/m^3 for refrigerant dryers and 8.6 W/m^3 to 50.6 W/m^3 for desiccant dryers (Langlais et al., 1991). These systems are not suited for isolated and remote communities.

Ozone generation with vacuum ultraviolet radiation (i.e. photochemical ozone generation) is ideal, as it requires no air drying system and can operate on ambient air (Eliasson & Kogelschatz, 1991; Mizuno et al., 2022; Salvermoser et al., 2009). Vacuum ultraviolet (VUV) radiation has sufficient energy (λ_{VUV} < 200 nm) to dissociate the dioxygen molecules (O_2) and produce ozone. Molecular nitrogen (N_2) does not absorb VUV radiation with wavelengths longer than 150 nm (Mizuno et al., 2022). The energy of photons with wavelengths above 127 nm is not sufficient to dissociate N_2 (Okabe, 1978). This dissociation is required for the formation of NO_x, which produces nitric acid in the presence of water vapor. Theoretically, VUV radiation does not generate nitric acid from humid gas. Two types of lamps are mainly used in VUV applications, low-pressure mercury lamps and excimer lamps (Alapi et al., 2018). Since the Minamata Convention in 2013, mercury-free alternatives have been sought to eliminate the risks associated with mercury on human and environmental health (Hsu et al., 2021; Sheikh et al., 2024). Xenon excimer lamps emit radiation with a wavelength of 172 nm and a full width at half maximum (FWHM) of 14 nm (Alapi et al., 2018; Mizuno et al., 2022). These lamps therefore constitute a mercury-free alternative for photochemical ozone generation.

The presence of water molecules in the carrier gas reduces ozone production by reacting with VUV radiation to produce OH and H radicals (equation 1). The H radicals are immediately converted to HO_2 (equation 2). The production of OH and HO_2 radicals promote ozone destruction reactions (equations 3 and 4). Equations 1 to 4 are taken from Salvermoser et al. (2009).

$$h\upsilon + H_2O \rightarrow H + OH \quad \text{(equation 1)}$$
$$H + O_2 + M \rightarrow HO_2 + M \quad \text{(equation 2)}$$
$$OH + O_3 \rightarrow HO_2 + O_2 \quad \text{(equation 3)}$$
$$HO_2 + O_3 \rightarrow HO + 2\,O_2 \quad \text{(equation 4)}$$

Taking into account these premises, a photochemical ozone generator using a xenon excimer VUV lamp (λ = 172 nm) was built. The generator was air-fed and powered by a 24 V source. The aim of this study was to evaluate the production capacity of a photochemical ozone generator fed with ambient air without a gas dryer, in order to

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decentralize ozone production. The effect of relative humidity in air on photochemical ozone generation at different gas flow rates was investigated.

METHODOLOGY

Experimental setup

The system presented in uses a xenon VUV lamp (*Radium XERADEX® L32*) to generate ozone from extra-dry bottled air (CAS : 132259-10-0). The lamp has an irradiation length of 250 mm and a diameter of 32.5 mm. A PVC tube (diameter = 50.8 mm, length = 450 mm) surrounds the lamp to seal the system and transport the carrier gas in the photoreactor region. A fan operates during all tests to cool the system. Valves control the relative humidity of the gas by directing dry air into a cylinder containing distilled water.

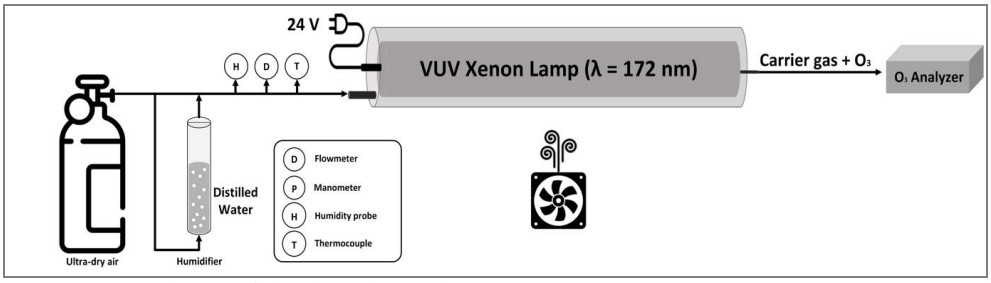

Fig. 1. *Experimental setup of the photochemical ozone generation system with the XERADEX® L32 lamp*

Analytical methods and equipment

The relative humidity (%) in air is measured using a Honeywell HIH-4000-001 humidity probe. Gas flow rates between 0.3 and 4.0 L/min are measured using a Dwyer VFB-65-BV flowmeter, and flow rates between 4.0 and 10.0 L/min are measured using a Dwyer FL-2014 flowmeter. The ozone concentration in the process gas is measured using UV photo-absorption with a Teledyne ozone analyzer (model 465H). The nitrate concentration (mg NO_3^--N/L) is measured using the UV spectrophotometric method (i.e. method 4500-NO_3-B in APHA (2023)) with the Agilent Cary 60 UV-VIS spectrophotometer.

Effect of relative humidity in air on photochemical ozone production

Preliminary ozone production tests have shown that the ozone concentration produced by the photochemical generator stabilizes after 20 minutes. For each test with variable humidity in air, the gas flow rate is set to between 0.3 and 4.0 L/min. The lamp power can be set to either 20 W or 35 W. All tests are carried out at a constant gauge pressure (P_{in} = 69 kPa, P_{out} = 0 kPa).

The relative humidity of the air is adjusted and fixed at values of 0 %, 20 %, 40 %, 60 %, 80 % or 100 % by means of a valve that controls the proportion of gas flow sent to the humidifier. Once the humidity sensor indicates a stable value (± 0.1 %) for five minutes, the lamp is activated for a 25-minute test. The ozone production value is determined from the ozone concentration after 25 minutes along with the gas flow rate (see equation 1). Bubbling tests of ozone-containing gas in a 100 mL solution of 0.1 M NaOH were carried out to monitor nitric acid production at 0 % and 100 % relative humidity.

$$Ozone\ production\ (g-O_3/h) = \frac{C_{O_3} * Q_{gas} * 60}{1000} \qquad \text{(equation 5)}$$

where:

C_{O_3} : Ozone mass concentration after 25 minutes of operation (mg-O_3/L_{gaz})

Q_{gas}: Volumetric gas flow rate (L/min)

RESULTS

Photochemical ozone generation from dry air

Fig. *2* shows the effect of gas flow rate on photochemical ozone production from dry air. The two curves illustrate the impact of lamp power (20 W and 35 W) on ozone production.

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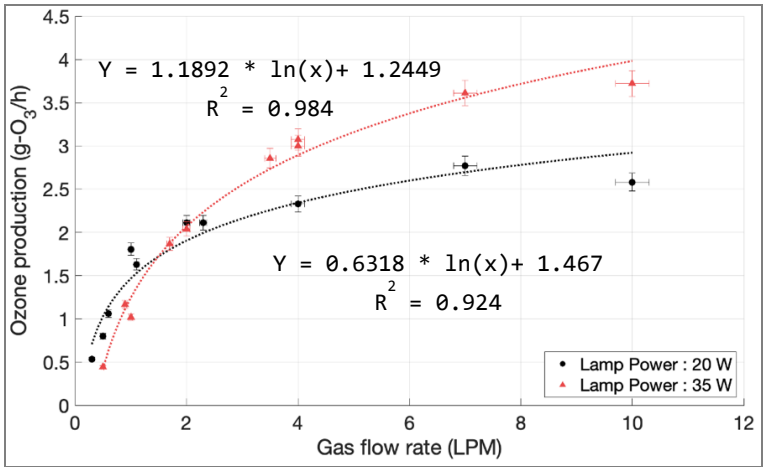

Fig. 2. *Photochemical production of ozone from dry air with a VUV xenon lamp (λ = 172 nm)*

The maximum ozone production is 2.8 ± 0.1 g-O_3/h and 3.7 ± 0.1 g-O_3/h for lamp power levels of 20 W and 35 W, respectively. These production quantities represent specific energy consumption values of 7.1 kWh/kg-O_3 (lamp power: 20 W) and 9.4 kWh/kg-O_3 (lamp power: 35 W). The gas produced at maximum production output has a mass concentration of around 6 mg-O_3/L. The maximum concentrations obtained in tests are 30 mg-O_3/L at 20 W and 17 mg-O_3/L at 35 W (Q_{gas} < 2.0 L/min). Ozone production is highest at 20 W lamp power for gas flow rates below 2.0 L/min (Fig. *2*). For higher gas flows (2.0 L/min < Q_{gas} < 10.0 L/min) the lamp set at 35 W produces more ozone.

Effect of relative humidity in air on photochemical ozone production

Fig. ***3*** shows the effect of relative gas humidity on ozone production from air. The curves represent different gas flow rates at equal pressure. As illustrated in Fig. *2*, an increase in gas flow positively impacts ozone production.

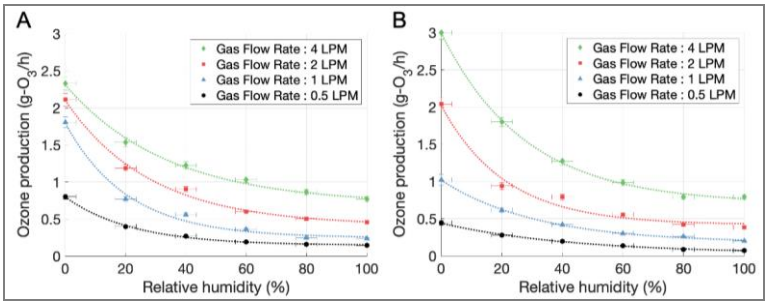

Fig. 3. *Effect of relative humidity in air on ozone production with a VUV xenon lamp (λ = 172 nm) set at power levels of [A] 20 W and [B] 35 W*

Increasing the relative humidity of the gas from 0 % to 20 % reduces ozone production by 35 % to 57 % for the lamp set at 20 W. A similar reduction in ozone production occurs for the 35 W lamp (37 % to 54 %). As the relative humidity rises from 20 % to 100 %, the ozone output of the 20 W lamp plateaus at a non-zero value for each gas flow rate. These values range from 0.15 g-O_3/h to 0.79 g-O_3/h (Fig. *3* A). For the lamp set at 35 W, ozone production values at 100 % relative humidity are similar to those of the 20 W lamp and range from 0.07 to 0.79 g-O_3/h (Fig. *3* B). Therefore, the maximum specific energy consumption values from water-saturated air (RH = 100 %) are 25.3 kWh/kg-O_3 et 44.2 kWh/kg-O_3 for the lamp set at power levels of 20 W and 35 W, respectively.

Bubbling tests in a NaOH solution were carried out on the ozone-containing gas produced by the xenon VUV lamp to determine the presence of nitric acid during ozone generation from humid gas. The results showed no nitric acid production at either 0 % or 100 % relative humidity, confirming the absence of NO_x production from VUV radiation.

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DISCUSSION

Photochemical ozone generation from dry air

The photochemical ozone generator has maximum ozone output rates of 2.8 ± 0.1 g-O_3/h and 3.7 ± 0.1 g-O_3/h for power levels of 20 W and 35 W, respectively. As the gas flow rate increases, ozone production rises until a plateau is reached. This effect is similar to the impact of gas flow rate on ozone production of dielectric barrier discharges (DBD) ozone generators (Boonduang et al., 2012; Cuong et al., 2019).

Thompson et Drago (2015) report that, on average, drinking water treatment plants in North America use 4.4 mg-O_3/L to treat surface water and 11.0 mg-O_3/L to treat groundwater. Indeed, groundwater requires higher ozone dosage for color abatement and the oxidation of organoleptic compounds in order to meet the treatment objectives (Thompson & Drago, 2015). Based on these conservative figures, the photochemical ozone generator set to 20 W has a surface water treatment capacity of 636 L/h and a groundwater treatment capacity of 255 L/h. When set to 35 W, the system's treatment capacity is 841 L/h for surface water and 336 L/h for groundwater. The Office of the High Commissioner for Human Rights (OHCHR) recommends a supply of 50 to 100 liters per person per day to meet basic health needs (OHCHR, 2010). Therefore, the production capacity of the photochemical ozone generator developed in this study is sufficient to produce treated water directly at the point of consumption (i.e. point-of-use production system) for a household.

The generator's ozone output corresponds to specific energy consumption values of 7.1 kWh/kg-O_3 (lamp power: 20 W) and 9.4 kWh/kg-O_3 (lamp power: 35 W). These values are slightly higher than the theoretical value of 6.7 kWh/kg-O_3 calculated by Salvermoser et al. (2008). In comparison, air-fed DBD ozone generators consume approximately 20 kWh/kg-O_3, whereas oxygen-fed systems consume between 8 and 13 kWh/kg-O_3 (Gottschalk et al., 2010). However, new developments in DBD ozone generation technologies that use water as ground and high-voltage electrodes enable specific energy consumption values of between 5.0 kWh/kg-O_3 to 9.4 kWh/kg-O_3 to be achieved (Xie et al., 2024; Xu et al., 2025). These new technologies therefore have comparable specific energy consumption values to the proposed photochemical ozone generator. However, these DBD ozone generators still require air drying or supply of bottled dry gas.

Photochemical ozone generation from humid air

In order to assess the possibility of feeding the photochemical ozone generator with ambient air, the effect of the relative humidity of the gas on the ozone production was determined. Initial tests confirmed the absence of nitric acid production in the gas generated by the photochemical ozone generator from humid air. This verification is crucial for validating the decentralized application of this type of generator. In contrast, the main issue with DBD ozone generation from humid air is the production of NO_x, which produces nitric acid when in contact with water vapor (Gottschalk et al., 2010; Rakness, 2015). Nitric acid damages the device and poses risks to human and environmental health (Rakness, 2015; World Health Organization, 2022). Therefore, DBD ozone generators require moisture-free gas, which can be supplied by cylinders or a gas drying system. These gas dryers consume between 5.5 W/m^3 and 50.6 W/m^3 of energy (Langlais et al., 1991).

The presence of water vapor significantly affects the ozone production of the system set at both 20 W and 35 W, reducing it by between 35 % and 57 %, as soon as the relative humidity increases from 0 % to 20 %. This reduction in production is primarily due to the dissociation of water molecules caused by VUV radiation, which favors the generation of OH and HO_2 radicals (see equations 1 and 2). These radicals are involved in reactions responsible for ozone destruction (Salvermoser et al., 2009). When fed with water-saturated air (RH = 100%) the photochemical ozone generator set to 20 W produces gas with ozone concentrations ranging from 0.15 g-O_3/h to 0.79 g-O_3/h. According to the average ozone dosage values provided by Thompson et Drago (2015), the photochemical ozone generator fed with water-saturated air can treat 180 L/h of surface water and 72 L/h of groundwater.

When fed with water-saturated air, the photochemical ozone generator set to 35 W does not produce more ozone than the system set to 20 W. These productions correspond to specific energy consumption values of 25.3 kWh/kg-O_3 for the 20 W lamp, compared to 44.2 kWh/kg-O_3 for the 35 W lamp. Therefore, the additional energy supplied by the 35 W lamp contributes equally to both the photochemical ozone generation and the destruction reactions through the production of OH and HO_2 radicals. Therefore, if the air has relative humidity values of more than 40%, it is preferable to set the lamp to 20 W.

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Decentralization of drinking water production

Centralized drinking water production systems are not suitable for rural areas. Transporting water over long distances and storing it for extended periods of time facilitate the growth of pathogenic microorganisms that cause waterborne diseases (Balasooriya et al., 2023; Rusca et al., 2022; Wright et al., 2018). In order to prevent the proliferation of these microorganisms prior to use, it is necessary to disinfect the water directly at the point of consumption. Technological solutions that accelerate the decentralization of oxidation and disinfection processes must avoid the dependence on the transport and storage of chemical products (Von Gunten, 2018). In light of this, the photochemical ozone generation, which is fed by ambient air and powered at a voltage of 24 V, aligns with these objectives. The system is indeed freed from the constraints associated with DBD ozone generators, such as the need for a high-voltage power supply and an air dryer or desiccants. Furthermore, the 24 V power supply enables the system to run on renewable energy while producing sufficient ozone to meet the basic disinfection needs of a household.

In order to provide resilient water treatment, decentralized drinking water systems must not depend on a single treatment process. The multi-barrier approach must be adopted to effectively remove diverse pathogens and toxic contaminants (Chen, Loeb, et al., 2017; Hogard et al., 2024; Pooi & Ng, 2018). In order to design a decentralized drinking water production system, the proposed photochemical ozone generator must be combined with filtration and adsorption modules. Decentralizing drinking water production will help overcome the challenges associated with the spread of waterborne diseases in rural areas.

CONCLUSION

The evaluation of a photochemical ozone generator with a xenon excimer VUV lamp (λ = 172 nm) indicated that as the gas flow rate increases, ozone production rises until a plateau is reached. Increasing the relative humidity of the gas from 0 % to 20 % reduces ozone production by 35 % to 57 % for the lamp set at 20 W. A similar reduction in ozone production occurs for the 35 W lamp (37 % to 54 %). As the relative humidity rises from 20 % to 100 %, the ozone output of the 20 and 35 W lamp plateaus at a non-zero value for each gas flow rate. When fed with water-saturated air, the photochemical ozone generator set to 35 W does not produce more ozone than the system set to 20 W. If the air has relative humidity values of more than 40%, it is preferable to set the lamp to 20 W. The absence of nitric acid in the gas produced from humid air validates the use of the photochemical ozone generator in a decentralized drinking water production system. It avoids the need to install an additional gas drying system, which would consume additional energy. Furthermore, as the system operates on a 24 V power source, it can be powered with renewable energy. Photochemical ozone generation is an ideal disinfection process to consider in order to accelerate the decentralization of drinking water production.

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AUTHOR'S CONTRIBUTION

Étienne Bérubé: Lead author. Responsible for the measurements on the structures. Data collection, formal analysis, and drafting of the article (review and editing).

Robert Hausler: Responsible for carrying out measurements on the structures. Manager of the structures for the study. Performer of measurements, conceptualization, data processing, formal analysis. Article writing (review and editing).

Mariana Acosta Lopez: Author. Performed statistical modeling. Created tables and graphs. Updated and final revised the article.

En este artículo no existen conflicto de interes entre los autores.

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