



Research article

BT100, a three-in-one, multipurpose disinfecting, deodorizing, and air-cleaning solution with an effective, gradual, and continuous gaseous chlorine dioxide-releasing substance

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ABSTRACT

Aerosols carrying viruses that are released from the oral cavity of infected individuals are the primary, if not the only, means of transmission during viral respiratory disease epidemics. This makes crowded rooms and tiny, enclosed public areas like bathrooms prime environments for the transmission of diseases. Volatile organic compounds (VOCs) and formaldehyde are two contaminants that pose serious threats to human health and well-being in indoor environments. The varied disinfectant properties of chlorine dioxide (ClO₂) make it a key player in treating a range of air quality issues. To balance effectiveness and safety, however, the careful application of chlorine dioxide is essential to achieving the best results in air quality while preserving human health and well-being. This study explores the many functions of chlorine dioxide, including the prevention of the spread of viruses, the elimination of harmful gases like ammonia and hydrogen sulfide, and its effects on formaldehyde and total volatile organic compounds (TVOCs) in indoor environments using BT100. The results indicate a reduction of 98.5%, 81.01%, 62.22%, 46.5%, and 63.84% in minimizing aerosolized viruses, ammonia, and hydrogen sulfide gas in addition to formaldehyde and total volatile organic compounds.

1. Introduction

Several of the deadliest epidemics and pandemics in the past 100 years have been caused by respiratory disorders, which are a major cause of death. These include the Spanish flu, which killed between 50 and 100 million people, the SARS-CoV pandemic of 2003, which killed 700 people in 37 countries, the Influenza-A pandemic of 2009–2010, which claimed over 28450 lives, the severe flu pandemic of 2013, and the COVID-19 virus, which infected over 758.3 million people [1–3]. Along with the human toll, these pandemics have significant economic costs. For example, the cost of the 2013 severe flu pandemic was predicted to be up to US\$ 3 trillion, or nearly 5% of the global GDP. For the COVID-19 pandemic, the Asia Development Bank predicts costs of US\$ 2–4.1 trillion.

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(2.3–4.8% of the global GDP) [2,4]. Given that airborne infectious microorganisms, that cause respiratory infections, are frequently found outside of medical facilities and pose a serious threat to human health [5], extra care must be taken to prevent or control the spread of airborne infectious diseases and improve indoor air quality [6–8]. To stop the possible spread of pathogens (bacteria and viruses) that might create a pandemic, it is crucial to adopt the health protocols advised by the government and use disinfectants, mainly air disinfectants. According to research by Ref. [7] all harmful microbes may live in the environment for hours to days. Today, it is widely acknowledged that one of the primary methods of airborne viral transmission involves inhaling bioaerosols produced by breathing, coughing, speaking, and sneezing [9,10]. The amount and size of possibly virus-containing particles that a person produces are highly influenced by their respiratory rate and amount of physical activity therefore, reducing the number of microorganisms floating in the air will be an effective way to minimize the occurrence of respiratory infections. In theory, a liquid droplet with a diameter of 100 μm would reach the earth within a few seconds if it were released from the respiratory system at 1.5 m above the ground. In the air, however, water evaporation causes the exhaled droplets to typically diminish quickly. Smaller particles are created as a result, and they can linger in the air for many hours [11,12]. In highly populated areas where many people are generally packed into small spaces, such as waiting areas at stations, the interiors of buses and trains, and public restrooms, there is a higher risk of infection. Indoor air quality (IAQ) can be improved by reducing emissions, good ventilation, and using air purification equipment [13–16]. Currently, the primary methods of indoor air purification include filtration [17], electrostatic [18,19], photocatalytic [20], and negative ion purification [21]. Filters are the most common kind of air purifier, accounting for over 90% of the market. According to Ref. [22] non-woven fiber-based high-efficiency particulate air (HEPA) filters are 99.97% efficient in removing small particles from the air. However, it is vital to change the filter medium often since the captured bacteria or particles would accumulate on the filter's surface. The pressure drastically decreases if the filter is not changed [23]. Additionally, the pollutant shedding will result in secondary pollution of fine particulate matter and bioaerosol in indoor air [24].

The potential transmission of diseases (bacteria and viruses) can also be stopped by air disinfectants. A disinfectant is a substance that is used to prevent or eliminate germs (such as bacteria, viruses, and fungus, except bacterial spores) on inanimate surfaces including furniture, rooms, floors, and others [8,25]. Since there is a chance that disinfectants can irritate the skin and perhaps cause cancer, they should not be applied to the mucous membranes or skin. This is distinct from antiseptics, which are used to disinfect mucous membranes and the skin's surface. Chlorine dioxide (ClO_2), is a member of the chlorine class of chemicals that includes sodium hypochlorite and hypochlorous acid that may kill viruses by penetrating their outer walls and damaging their interiors [8, 25–29]. With a molecular weight of 67.45 g mol^{-1} and a yellow-green color at low concentrations, chlorine dioxide is a water-soluble gaseous compound. There is little reaction byproduct and minimal reactivity with organic materials. The FDA approved 5–450 ppm for foods such as fruits and vegetables in 1995, while the EPA allowed the concentration of dissolved chlorine dioxide in drinking water to be less than 0.8 ppm. It also has good sterilizing capability [8,25–29]. Chlorine dioxide is estimated to directly oxidize proteins containing tyrosine, methionyl, or cysteine, which damages the structurally important regions of the enzyme or membrane protein involved in metabolism [26–30]. The primary sterilization mechanism easily oxidizes the amino acids cysteine, tryptophan, and tyrosine, but not to the RNA of the virus [26,30–32]. The secondary sterilization mechanism was for physiological functions and hindered protein synthesis [28,29,33], but recent studies have also shown that it hinders the permeability of the outer membrane [31–34]. Such chlorine dioxide is known to be more effective in controlling microorganisms because it is more permeable in the form of gas than in the form of liquid chlorine dioxide [35].

Chlorite and water can be electrochemically reacted to generate chlorine dioxide gas but this cannot be used in toilets or tiny areas, or chlorine dioxide can be made using a chlorine dioxide generator system. To solve this problem, we created the BT-100, a simple-to-use disk with a breathable film that gradually releases chlorine dioxide gas by adjusting the quantity of chlorine dioxide-generating material, reaction, and reaction time of chlorine dioxide. This study confirmed the effect of chlorine dioxide inactivating the MS2 virus, one of the index microorganisms in the air, in a space resembling a restroom on a train. Additionally, we tested its effect on ammonia and hydrogen sulfide, TVOC, and HCHO which are odor and harmful sources. In metropolitan locations, inappropriate maintenance of public facilities, such as restrooms at movie theaters, bus/railway stations, hospitals, retail centers, etc., produces unpleasant odors that have an impact on both users and nearby neighbors. Congested marketplaces prevent odor from leaving them, which is problematic for both store owners and shoppers, so it is expected that this study will be used as a basis for developing space sterilization systems linked to HVAC and IAQ in the train in the future.

2. Materials and methods

2.1. Preparation of host and virus

Model systems can serve as a proxy for human enteric viruses while having no negative impact on health, making them an effective testing tool. The MS2 bacteriophage, commonly utilized as a viral model system [34,36,37], has been demonstrated to be acceptable for this investigation. Bacteria (*Escherichia coli*, ATCC 15597) and bacteriophages (*Escherichia coli* bacteriophages MS2, ATCC 15597-B1, Manassas, VA, USA) were employed as the model system in this work. The surface of the plates was gently cleaned with sterile distilled water followed by a 24-h incubation period. After incubation, the plates' surface was gently wiped with pure distilled water. According to the ATCC product sheet, the host was grown. The 1000 μL stock solution of the MS2 bacteriophage was frozen (-75°C) for future research, and a few stock solutions were cultured with bacteriophages to test their vitality. A 40 mL sterilized sample of tryptase soy broth was used to create the stock solution of *E. coli* C3000 (1000 μL) for the tests. This mixture was put into a shaking incubator at 37°C and 150–200 rpm. The bacteria were cultivated overnight (6–12 h). The stock solution (1 mL) and 50 mL of sterile deionized water were combined to create MS2 bacteriophages. Additionally, MS2 stock solutions were made in compliance with

the product sheet provided by the American Type Culture Collection (ATCC). The MS2 was then aerosolized using an atomizer connected to a diffusion drier (silica gel tube) to absorb moisture. A mass flow controller (MFC) was used to pump, clean, and regulate compressed air. This was attached to the atomizer, and the aerosolized virus solution was fed into the test chamber through a 4×4 channel. Bioparticle counter MBio 150 device (Mediaever Co. Ltd.) was used to measure the amount of aerosolized virus.

2.2. BT100 product

The BT100 product is a chlorine dioxide sustained-releasing disk that may submerge the precursor capable of producing chlorine dioxide while steadily releasing chlorine dioxide gas. Aluminum silicate (83–88%) serves as the primary carrier for the mixture, along with citric acid (<3%), carboxymethylcellulose (5–10%), sodium chlorite (<2%), and water (<2%). The Water Vapor Transmission Rate (WVTR) moisture permeability ranges from 5000 to 8000 mg/24 h at 60% relative humidity. The product is covered with a lid film composed of a high-base weight material like PVC or PP that is not air permeable after being sealed with a multi-film. When the lid layer is removed again, the disk, which is a moisture-reactive chlorine dioxide producer, collects moisture from the air and sets off a reaction. The amount of ClO_2 gas generated at this time was measured by GAS TIGER 2000_ ClO_2 Tester (Wandi, China), and the amount of chlorine dioxide generated in the normal state is as follows. Scanning electron microscopy was used to examine the surface morphologies of the BT100 disk (SEM-Hitachi SU-70, Japan).

2.3. Experimental chamber

A $1 \times 1 \times 1 \text{m}^3$ square experimental chamber built of steel/FRP double layers with windows attached to the front for measurement and a 10 cm hole drilled on the opposite side to connect duct for viral setup and sterilizing experiment was built. The size of the chamber was maintained at this level because it was not appropriate as an experimental method in a small space like a train toilet. The chlorine dioxide release disk used for sterilization and deodorization tests was tested using a product with a capacity of 40 g/pcs. A tiny fan was also installed inside the chamber to allow air to flow to one side for 1 m and 5 s and air to the other side (see Fig. 1). The BT100 disk was 50 cm above the chamber's floor in the middle. Experiments for testing the sterilization effect were performed using the MS2 virus whereas all the experiments for deodorization were done using ammonia gas and hydrogen sulfide gas. All the experiments using MS2, ammonia (NH_3), and hydrogen sulfide gas (H_2S) were performed using the same setup.

For standard measurement first, the concentrations were measured without using the BT100 inside the chamber. After 20 min, the BT100 device was added inside the chamber followed by adding aerosolized MS2, NH_3 , and H_2S gas. To aerosolize the MS2, a diffusion drier (silica gel tube) was coupled to an atomizer (model 9302, TSI Inc., Shoreview, MN, USA). The particle generation capacity of the atomizer was more than 10^7 particles/ cm^3 with a flow rate of 6.5 L/min. A mass flow controller (MFC) was used to pump, clean, and regulate the compressed air. The aerosolized virus solution was then fed through a 4×4 duct configuration attached to an atomizer inside the chamber (Fig. 1). The MBio 150 device (Mediaever Co. Ltd.) was used to measure the amount of aerosolized virus with and without the BT100.

For testing the deodorizing property, ammonia gas was using Ammonia solution (Sigma-Aldrich Extra pure $\geq 99.98\%$, 35%, 4 M in methanol), and H_2S was prepared by reacting NaHS with sulfuric acid ($2\text{NaHS} + \text{H}_2\text{SO}_4 \rightarrow 2\text{H}_2\text{S} + \text{Na}_2\text{SO}_4$) were diluted to 100 ppm each and used. The measuring device was the GAS TIGER 2000_Multi sensor Tester. First, these were injected inside the chamber without the BT100 product for 30 min to obtain a standard concentration, and later on the concentration was measured after keeping the BT100 product for up to 9 h to check the results after slow and sustained release of ClO_2 .

3. Results and discussion

3.1. BT100 product

Additionally, the chlorine dioxide sustained release disk (Brand: Cloon BT100s), which can release chlorine dioxide gas gradually,

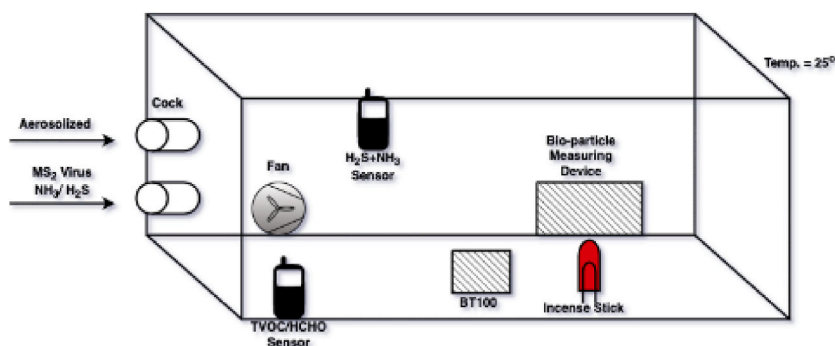


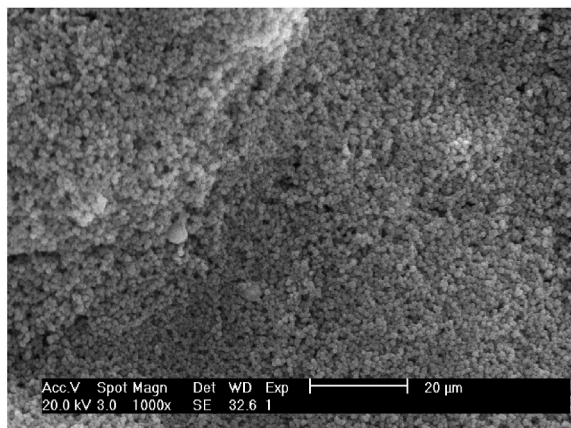
Fig. 1. Diagrammatic representation of the experiment using BT100 product.

Table 1

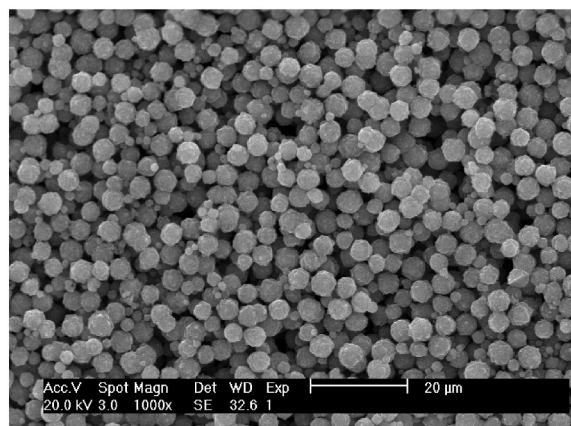
Release amount of chlorine dioxide from BT100 product.

40gsm Disk	Release rate (day) @20 °C, 65RH																							Total release amount (ppm/47days)	
	1	3	5	7	9	11	13	15	17	19	21	23	25	27	29	31	33	35	37	39	41	43	45		47
ppm	0.3	0.7	0.8	0.7	0.7	0.7	0.7	0.6	0.6	0.5	0.5	0.4	0.4	0.4	0.4	0.4	0.4	0.4	0.4	0.3	0.3	0.3	0.3	0.3	11.5

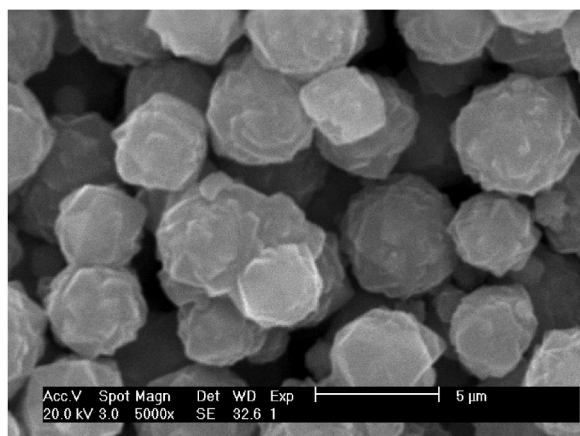
mixes the precursor capable of producing chlorine dioxide with various materials before immersing it in a porous crystalline aluminum silicate carrier. After drying the carrier, the WVTR moisture permeability is 5000–8,000mg/24hr, 60%RH. It is a product that is sealed with a multi-film and then covered with a lid film made of a high-base weight material such as PVC or PP that is not air permeable. The disk is a moisture-reactive chlorine dioxide generator that absorbs moisture in the air when removing the lid film and causes a reaction. The amount of chlorine dioxide gas generated at this time was measured by GAS TIGER 2000. ClO₂ Tester (Wandi, China), and the amount of chlorine dioxide generated in the normal state is shown in Table 1. The structural results obtained from the BT 100 product



(a)



(b)



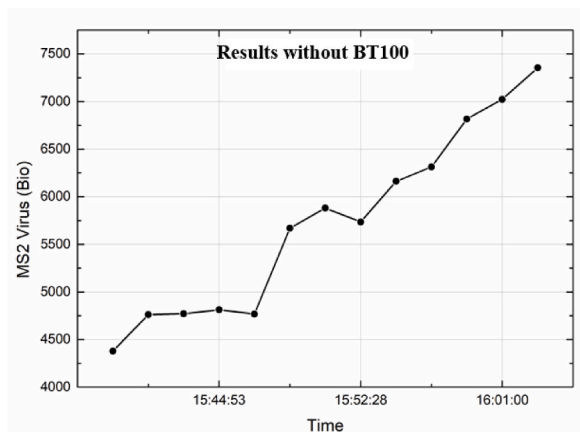
(c)

Fig. 2. (a,b,c)- Scanning Electron Microscopy results of the BT100 product.

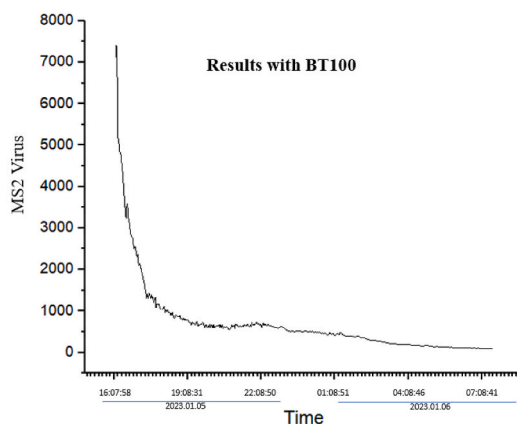
through SEM are shown in Fig. 2.

3.2. Antiviral activity test of BT100

Chlorine dioxide (ClO_2) has a strong oxidization capability even in acidic environments, making it one of the most effective disinfectants to date [38,39]. According to Ref. [40] chlorine dioxide has a solubility five times more than that of chlorine and an oxidation capability 2.63 times greater than that of chlorine gas. It eliminates the protein anabolic pathways, killing all types of microorganisms, including spores, bacteria, viruses, fungi, and Clostridium botulinum [41–43]. Chlorine has the advantage of cheaper costs of operation and preparation but is difficult to store or transport due to its chemical structure [40,44]. ClO_2 is yellowish gas at room temperature and is relatively easily dissolved in water [solubility in water is 8 g/L (20 °C and 70–100 mmHg) [38]. However, since ClO_2 gas dissolved in water dissipates within a short period of time, the dissolved level of ClO_2 gas decreases as time passes. Consequently, ClO_2 solution has been used by generation on site. ClO_2 is an oxidizer, which is reduced to chlorite ion (ClO_2^-) by capturing an electron ($\text{ClO}_2 + e^- \rightarrow \text{ClO}_2^-$). The redox potential (E°) is relatively high as 0.95 V ClO_2^- , in the presence of water, captures four electrons and is reduced to chloride ion (Cl^-) ($\text{ClO}_2^- + 2\text{H}_2\text{O} + 4e^- \rightarrow \text{Cl}^- + 4\text{OH}^-$), whose E° is 0.78 V which is lower than that of ClO_2 . Consequently, the oxidative activity of ClO_2 is stronger than ClO_2^- . The E° of ClO_2 is lower than that of hydroxyl radical ($\cdot\text{OH}$, 2.8 V), ozone (O_3 , 2.07 V), and hypochlorous acid (HClO , 1.49 V), therefore, it undergoes more selective oxidative reaction than these other molecules [34,45]. ClO_2 has a very broad range of applications since it may be employed as a gas or a solution. ClO_2 has been used for bleaching paper pulp for a very long period, mostly in Western nations [8]. Among the numerous types of carbohydrates present in the pulp, ClO_2 , which is distinct from Cl_2 , preferentially oxidizes lignin [36,37]. Comparing ClO_2 to other chlorine-type bleaching agents, it also has the benefit of minimizing the formation of dioxins. Therefore, ClO_2 acts as an alternative to the bleaching by Cl_2 since it is ECF (Elemental Chlorine Free) and doesn't produce dioxins. The U.S. Food and Drug Administration (FDA)



(a)



(b)

Fig. 3. (a,b)- Sterilization test (using aerosolized MS2) results with and without BT100 product.

also permits the use of ClO_2 as a food additive for bleaching cereal flours and sanitizing produce. Additionally, the United States Environmental Protection Agency (EPA) has approved the use of ClO_2 as a disinfectant for tap water (less than 0.8 ppm for targeted water quality) [46], and the molecule is used at water treatment facilities in the U.S.A. as a disinfection method that produces little trihalomethane, a carcinogen. Another source for chlorine-containing disinfectants is sodium hypochlorite [40,47]. The use of sodium hypochlorite is characterized by relatively lower toxicity, simpler equipment, more stable operation, easier control, and lower operation and preparation costs when compared to other chlorine-containing disinfectants [47]. However, it should be noted that sodium hypochlorite disinfection uses more energy and produces more pollution and corrosiveness [48]. Therefore, a safe and dependable sodium hypochlorite generator with high electrical efficiency, low water consumption, low salt, and energy consumption, extended operational life, and simple operation should be used when employing on-site generated sodium hypochlorite for disinfection. Additionally, the EPA has approved the use of high concentrations of ClO_2 gas as a sterilizer for tools, clean rooms, environmental surfaces, and manufacturing and experimental equipment [45] more recently, Annex G of NSF/ANSI 49 standardized the gas as an alternative to primary fumigation for the sterilization of biosafety cabinets.

The findings of the current study are consistent with those of several studies that have characterized ClO_2 gas as a good disinfectant with a broad range of antibacterial activity [41–43]. We verified that the low concentration of ClO_2 gas prevented the infection with the MS2 virus [49]. We anticipate that in the future, legal laws will be created that can be used for both high- and low-concentration ClO_2 gas IAQ improvement. In the case of viruses that are difficult to cultivate due to the nature of the virus or that are difficult to experiment with directly due to high infectivity, an alternative virus is used. Bacteriophage is a type of virus that uses bacteria as host cells, so it can be easily cultivated in laboratories without cell culture facilities. Bacteriophage MS2 (ATCC 15597-B1) was purchased and used by ATCC, and the conditions in the chamber were 25 °C, 65% RH, air flow 1 m/5 s, and the experiment was blank and Cloon BT100 to check the change. Bacteriophage MS2 measurement was measured by MBio 150 device (Mediaever Co. Ltd). Fig. 3 shows the initial concentration of the 7353 particles whereas with time, due to the slow and sustained release of ClO_2 , the end concentration reaches 90. The overall average decrease in the aerosolized MS2 virus average concentration was found up to 98.58%. These results are similar to that of the findings of [50]. However, according to their research, the biocidal effect of ClO_2 was found to diminish with an increase in time after 10 min due to the observed tailing, which may have been acquired after the virus was synthesized by attachment

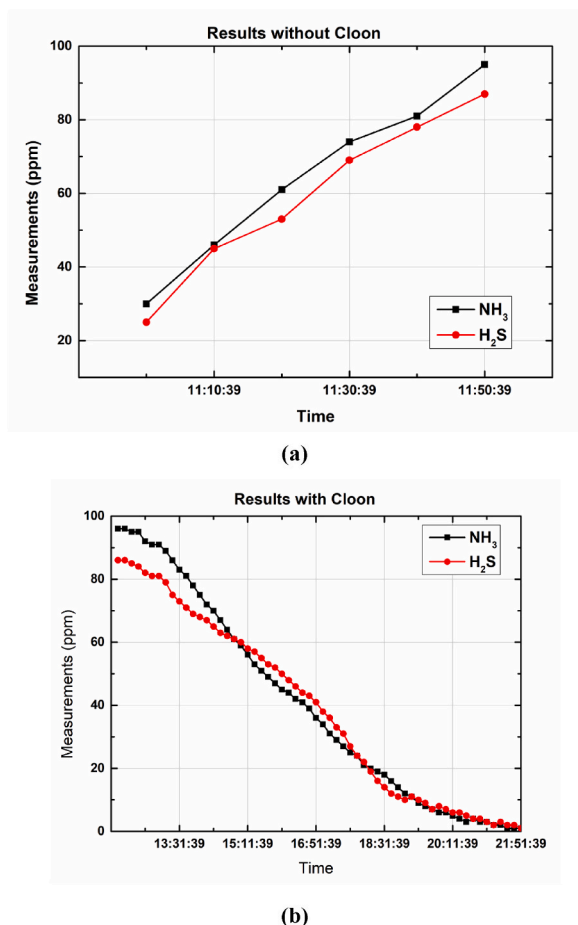


Fig. 4. (a,b)- Deodorization test (NH_3 and H_2S) results with and without BT100 product.

to other (virus)particles or as a result of intrinsic virus population heterogeneity whereas with our product this was not observed. According to earlier research [50,51], environmental surfaces, especially when moist, are important in the propagation of viral infections and act as a reservoir for different bacteria whereas with our product the moisture helps in slow release of ClO₂ due to which up to RH 65% the BT100 works perfectly.

3.3. Deodorization test and volatile organic compound reduction properties of BT100

Aside from the in-situ sewage treatment, the biggest drawback of the outdoor toilet is its odor. The smells connected with ammonia and hydrogen sulfide are thought to be the most repulsive of them. To date, several deodorization techniques have been created to solve this issue. Among these are sensory deodorization (36), chemical deodorization (37), which involves desulfurization and ion exchange, and physicochemical deodorization (35), which makes use of activated carbon or synthetic zeolites. There are benefits and drawbacks to each of these approaches, and there are still many obstacles to deodorization applications in everyday life. According to research, chlorine dioxide can successfully lower ammonia levels in enclosed spaces. When combined with ammonia, it creates chloramines, which are less flammable and smell more subtly. However, variables like the chlorine dioxide concentration, contact time, and beginning ammonia concentration may all have an impact on how well ammonia is reduced [52]. Hydrogen sulfide may be oxidized by chlorine dioxide, which then produces sulfates or other less pungent chemicals. Chlorine dioxide has been shown in studies to be efficient at lowering hydrogen sulfide levels and managing smells in enclosed spaces. The unpleasant smell is helped to neutralize and the air quality is improved by the oxidation process between chlorine dioxide and hydrogen sulfide [53]. Although chlorine dioxide can be useful in lowering ammonia and hydrogen sulfide levels, it should be used with caution to guarantee safety and prevent any potential health hazards. Strong oxidizer chlorine dioxide can be dangerous in excessive doses or if improperly handled. To

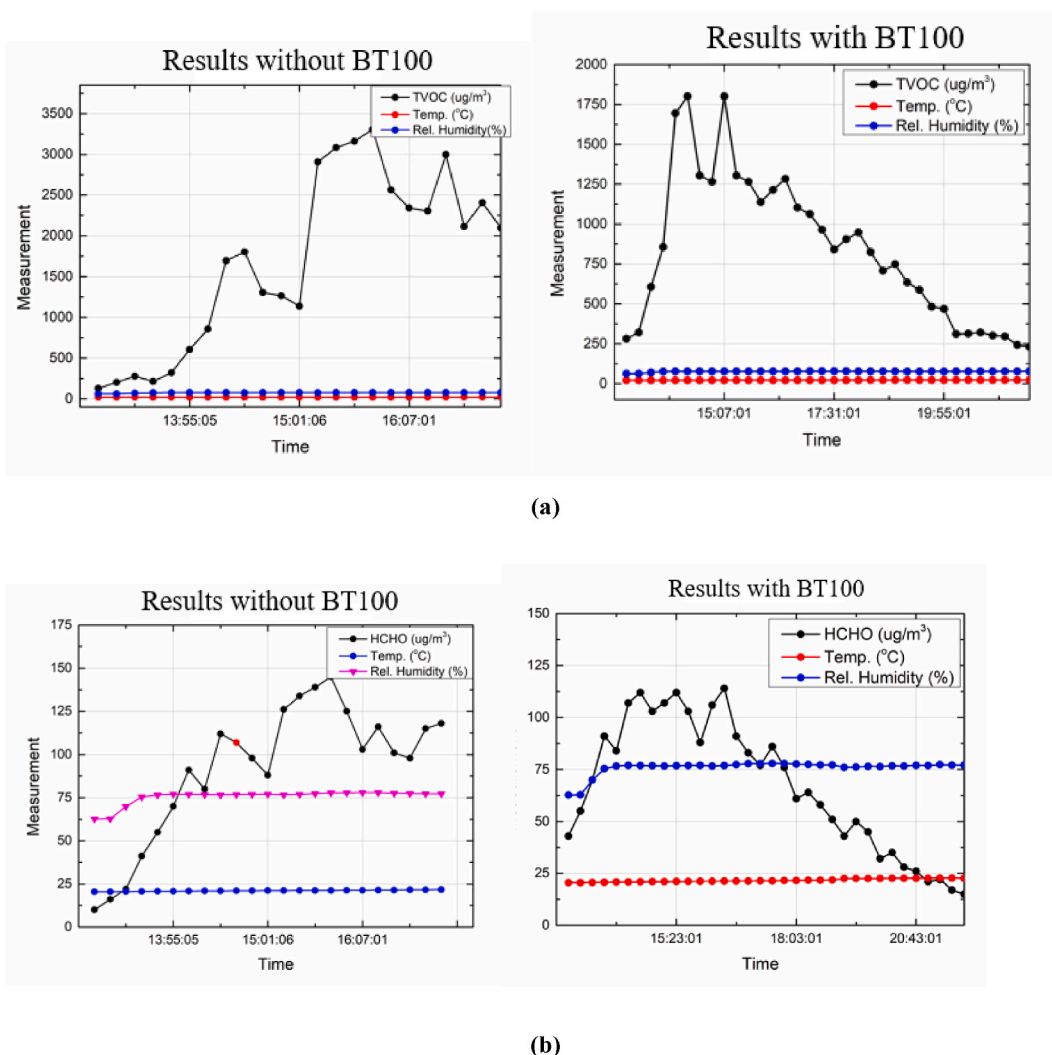
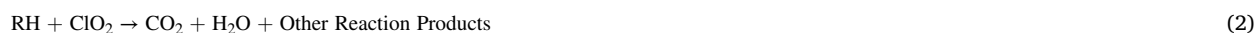


Fig. 5. (a,b)- TVOC and HCHO results with and without BT100 product.

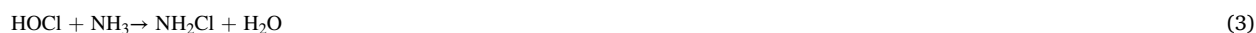
guarantee efficient odor control while limiting any potentially harmful effects, proper dose, application, and ventilation are crucial.

In this study, we performed a deodorizing experiment using ammonia and hydrogen sulfide using BT100. In Fig. 4(a and b), with BT100, we found a slow reduction in the ammonia and hydrogen sulfide gas with the passage of time. In the last 20 min s, the average reduction of ammonia and hydrogen sulfide was found to be 81.01% and 62.22%. We further confirmed the efficacy of this stable chlorine dioxide treatment to prevent the creation and growth of many unwanted VOCs based on the findings of the research done in Refs. [54,55] concerning the capacity of gaseous chlorine dioxide to decrease volatile organic compounds. When formaldehyde and VOCs are present, chlorine dioxide gives the organic molecules oxygen atoms or electrons, which causes them to disintegrate into simpler and less dangerous chemicals [Eqs (1) and (2)]. Instead of breaking the carbon-carbon and carbon-hydrogen bonds to form simpler, less volatile compounds and water vapours as it does with VOCs, chlorine dioxide reacts with formaldehyde molecules to convert them into carbon dioxide by oxidizing them and chloride ions. The following equations show the potential mechanism pathway.



We evaluated the BT100's effectiveness in reducing TVOCs and HCHO. Our findings in Fig. 5(a and b) demonstrate that BT100 is effective in lowering TVOC and HCHO by up to 63.84% and 46.5%, respectively.

Strong reactive species are produced by the ammonia/chlorination process and are in charge of degrading stubborn pollutants [52, 53,56]. According to Eqs [3–10], it has been hypothesized that dichloramine is unstable in neutral environments and that both chloramine and dichloramine break down into more reactive species such as radical OH, NOH/NO⁻, and ONOOH/ONOO⁻.



4. Conclusions

Emphasizing its relevance in improving air quality across several sectors are chlorine dioxide's multiple capacities in virus reduction, ammonia and hydrogen sulfide gas removal, and VOCs and formaldehyde mitigation. Chlorine dioxide is an effective instrument for improving health, safety, and environmental wellbeing due to its adaptability to many situations, quick action, and non-residual nature. Chlorine dioxide is a flexible ally in our pursuit of cleaner, safer, and more pleasant indoor places as our understanding of indoor air quality develops. While there are many advantages to using chlorine dioxide to enhance air quality, it is important to think about its correct and safe use. To optimize the advantages and minimize the risks of chlorine dioxide, it is essential to keep an eye on concentrations, abide by established exposure limits, and adopt industry best practices. With longer time intervals, our product, BT100, shows significant reduction results, suggesting that the pollutant concentration will also decrease and that ClO₂ gas release would occur more slowly. Our data indicates that there was a continuous release of gaseous chlorine dioxide that was regulated in compliance with USEPA regulations. The concentrations of aerosolized viruses, ammonia, hydrogen sulfide, formaldehyde, and volatile organic compounds were all found to have significantly decreased. The results show a reduction of 98.58%, 81.01%, 62.22%, 46.5% and 63.84% suggesting that this can work very well for small spaces such as subway washrooms, toilets inside trains, tiny indoor spaces.

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Data availability statement

Data will be made available on reasonable request.

CRediT authorship contribution statement

Sharma Shambhavi: Writing – original draft, Software, Methodology, Formal analysis, Data curation, Conceptualization. **Honggil Kim:** Writing – review & editing, Validation, Supervision, Resources, Investigation, Conceptualization. **Muhammad Jahanzaib:** Methodology, Investigation, Data curation. **Jooyeon Lee:** Methodology. **Duckshin Park:** Writing – review & editing, Supervision, Resources, Project administration, Funding acquisition.

Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Sharma Shambhavi reports financial support was provided by Korea Railroad Research Institute. Duckshin Park reports a relationship with Korea Railroad Research Institute that includes: employment. The authors declare no conflict of interest. If there are other authors, they declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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