Release of undesired by-products during the operation of virus inactivating air cleaning devices

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SUMMARY
Mobile air cleaner are seen as an effective tool for reducing airborne viruses in indoor air. The basis of the investigations was the determination of the reduction in virulence by means of inactivating technologies on basis of plasma- and UVC-technologies. The particle release of different particle fractions relevant to aerosols and the reduction of the virulence have been measured by injection of the non-hazardous surrogate viruses Phi6-bacteriophague with similar structure and environmental behaviour to SARS-CoV-2-virus into a climate-controlled test chamber. However, there are legitimate concerns regarding the release of harmful by-products, e.g. ozone or volatile organic compounds (VOC). In addition to the virulence tests the devices were investigated to the formation of such substances. The released substances were finally compared with limit values of national and international standards.

KEYWORDS
Virus, plasma technology, UVC technology, ozone, VOC

1 INTRODUCTION

The risk of infection by airborne viruses indoors can be reduced in three ways: dilution of the virus-containing aerosol by increasing the supply of fresh air, removal of the aerosol particles using filtering air purifiers, and inactivation of the viruses in the aerosol using irradiating air purifiers with cold plasma or UVC technology. We studied the efficiency of plasma- and UVC-technologies by measuring the reduction of virulence of a non-hazardous surrogate virus with similar structure and environmental behaviour to SARS-CoV-2-virus into a climate-controlled test chamber. This test method has meanwhile become established in as an expert recommendation for testing of filtering and inactivating air cleaning devices which require an investigation of the release of hazardous substances for inactivating devices (VDI-EE 4300-14, 2021).

The UVC range from 200 to 280 nm exhibits the highest antimicrobial and antiviral efficacy, as this wavelength hits the range where nucleic acids are particularly absorptive. The way UVC light works is by inactivating RNA through dimerization of two adjacent uracil bases. With focus on SARS-CoV-2 virus, the optimum of RNA absorption is between 240 and 280 nm with maximum absorption at 260 nm. For an effective reduction of corona virus activity above 90 % in air, radiation doses of 70 J/m² are necessary. An undesired by-product when using classical UVC emitters, is ozone. This can be generated by transmission of UVC wavelength lower than 210 nm (Kowalski, 2009; Batakliev et al, 2014; Heßling et al, 2020). The ozone producing wavelength can be inactivated by using special coated fused silica ("ozone-free emitter") which were doped with titanium dioxide or silica coating (Claus, 2021). An established preservative technique to induce microbial destruction on (food-) surface and water is formed by a plasma gas between electrostatic discharge between different

electrodes (Pankaj et al, 2018; Barjasteh et al, 2021). Plasma is defined as a physical gas state consisting of a mixture of neutral gas particles and a variety of charged particles and reactive components. Different electrical discharge variants are used as plasma source, such as corona, microwave, electrical arc and dielectric barrier discharge. Depending on the electro technical design, manufacturers offer these devices under the terms cold plasma, ionizers, non-thermal plasma or dialectical discharge. Plasma treatment on microorganisms leads to an immediate oxidative reaction on the cell membrane in bacteria and the protein capsid of viruses. DNA or RNA are destroyed by the attack of reactive components (Lai et al, 2016; Niedzwiedz et al, 2019).

Due to the exposure times and relatively low plasma temperatures, few undesired reaction products like volatile organic compounds (VOC) and ozone have been identified (Preis et al, 2013; Liao et al, 2017). There are legitimate concerns regarding the release of such harmful by-products. Recent studies about air cleaner based on photocatalytic reactions additionally indicate these findings (Gunschera et al, 2016; Salthammer et al, 2017; Joo et al, 2021). The European Centre for Disease Prevention and Control (ECDC, 2020) and the Commission on Indoor Air Hygiene at the German Federal Environmental Agency (Umweltbundesamt UBA, 2020) published statements for the use of indoor air purification technologies. They require the exclusion of non-hazard by-products during the operation of such devices. Taking these requirements in account, we tested commercially available air purifiers. Our study included investigations of virulence as well as the formation of undesirable by-products. The investigations were carried out under constant climatic conditions in a test chamber simulating a small classroom. The hypothesis was that the inactivating technology reduces load of active viruses in the room caused by an infectious student by reducing the virulence whilst a minimum of by-products are formed.

2 MATERIALS/METHODS

The indoor air cleaner to be examined were of higher price and quality from leading manufacturers available on the market. The selection was intended to cover a wide range of UVC- or plasma-technology, especially with and without filter. All selected devices were specially designed for virulence reduction. The technical specifications of the six investigated air cleaner are given in Table 1.

Table 1. Investigated air cleaner.

Device No [1]	Inactivating unit (additional filter technology)	Sound pressure level [db] [2]	Max. Power consumption [W] [2]	Measured flow rate [m³/h] [3]
	UVC-Technology:			
UVC_1	➢ UVC (without filter)	50	170	800
UVC_2	➢ UVC (coarse filter in front of the unit)	30 – 55	240	450
UVC_3	➢ UVC (coarse filter in front of and HEPA filter behind the unit)	25 – 45	110	1000
	Plasma Technology:			
CP_1	➢ Ionization (dialectical discharge without filter)	n. s.	25	400
CP_2	➢ Cold plasma (coarse filter in front of and behind the unit, activated carbon filter behind)	n. s.	n. s.	120
CP_3	➢ Cold plasma (coarse and HEPA filter in front of and activated carbon filter behind the unit)	30 – 50	75	400

[1] Anonymized in order not to draw any conclusions about specific manufacturers
[2] Manufacturers specifications (if available), rounded values, n. s. – not specified
[3] Adjusted and measured at the air outlet during measurements, rounded values

The investigations were carried out in the Fraunhofer IATC, a climate-controlled test chamber (Indoor-Air-Test-Center, round 130 m³, inert stainless steel, adjusted to 19 °C and 40 % RH, without external air flow). The interior was equipped with tables and chairs mimicking a small classroom. An aerosol generator (AGK 2000, Palas), was used to generate the aerosol containing Phi6-bacteriophague. Mixing in the room was ensured by two fans. After switching on the air cleaner the fans were switched off to guarantee that further mixing is exclusively provided by the device itself. The air flow was adjusted to ensure a theoretical air exchange of minimum 4/h. The devices were placed according to manufacturer's specifications (e.g. wall, ceiling, and ground). If necessary two or more devices were placed to ensure the theoretical air exchange. All particle counters (Fidas Frog, Pallas; P-Trak, TSI; WCPC 3788, TSI), the ozone analyser (O3 41M, ansyco), and VOC and airborn germ samplers (BiVOC2; Holbach); MBASS30, Holbach), were placed equidistantly to the aerosol generator and the air cleaner and recorded the entire testing runtimes.

Harmless Phi6-bacteriophagues with similar structure, size and environmental behaviour to the pathogenic SARS-CoV-2 virus used to perform the testing. They were freshly cultivated in host cells (Pseudomonas sp.) by plaque assay method before the chamber test. The suspension was continuously nebulized by the aerosol generator at $1.5 * 10^5$ Pa inlet pressure into the chamber atmosphere. After 1 h an atmosphere containing an aerosol with high viral load was generated. Now the air cleaner was switched on. Both aerosol generator and air cleaner runs for additional 1.5 h. Germ samplings on gelatine filter took place: half hour right before the devices switched on (P1), 30 min (P2) and 1 h (P3) after switching on the air cleaner. Subsequently the microbial analysis by means of a second plaque assay method were initiated (EN 13610, 2002; Zhao et al, 2014). The VOCs and selected carbonyls (aldehydes/ketones) were sampled: background ("blank") of the chamber, at the hour right before switching on the device, 5 min after switching on and continued for a further one hour. VOCs were trapped as double determination on Tenax TA®/Carboxen 1003 (Supelco) adsorbents and aldehydes/ketones on DNPH silica cartridges (Waters) for subsequent instrumental analysis (TD-GC-MS Markes TD 100 with Shimadzu GC 2010 and Shimadzu QP 2010 Plus, HPLC-DAD Agilent 1290 Infinity).

In addition to the virus tests, the air cleaner CP_1 using ionization technology (dialectical discharge) without any filters was additionally examined in an atmosphere with high VOC background load. The technology allows different ionization intensities, which was stepwise adjustable for the ionization levels 12.5, 25.0, 31.3 and 100 % of the maximum power consumption. A mixture of representative indoor air contaminants consisted of non-hazardous substances in same ration without carrier solvent was injected into the test chamber by a syringe pump: isobutanol, acetophenone, limonene, hexanal, and pentanoic acid. The injection started 1.5 h before the air cleaner was in operation in order to reach a constant concentration level within the chamber. Two final concentrations at 275 µg/m³ and at 850 µg/m³ were generated. The concentration was continuously monitored by a photoionization detector (PID, ppbRAE 3000, Honeywell). In sum 8 VOC test runs were conducted for each concentration step and each ionization level. VOCs and carbonyls were sampled as mentioned above with shorter sampling times and two subsequent sampling points after switching on the device. The installed sensors, detectors, and analyzers recorded the ozone and TVOC concentrations during the entire test runs.
All injection and sampling tests were carried in compliance with high safety measures and personal protective equipment for the staff in order to avoid particle and ozone inhalation. The sensors and detectors could be observed from the outside and the test runs external aborted if necessary.

3 RESULTS AND DISCUSSION

The testing of air cleaners for the efficiency to reduce the virulence of airborne viruses was meanwhile implemented in a VDI expert recommendation (VDI-EE 4300-14, 2021). The standard describes the injection of viruses to a maximum load before the device is running. As soon as the air cleaner is switched on the injection has stopped. To meet the requirements the viral load has to be reduced by one log-step (90 %) within half an hour. We could demonstrated it by validation measurements for a computing tool, which was developed in our institute (results also presented at the Indoor Air conference 2022 by Schmohl et al). At variance the investigations were not carried out with a spot injection of the virulent particles but with a continuous aerosol injected to reach a higher viral load as it occurs in case of a superspreader event.

Determination of the virulence

For the virus injection the harmless Phi6-bacteriophagues were applied as a realistic model virus for the pathogenic SARS-CoV-2, which is only allowed to be handled in a high security laboratory protection level 3, in order to avoid hazards for staff and environment. In general, Phi6-bacteriophague has already been used as a suitable surrogate for coronaviruses for numerous different studies because of its similar structure and size and its almost identical environmental behavior (Fedorenko et al, 2020; Ling et al, 2020). Table 2 shows the virulence in % of the Phi6-bacteriophages in the test chamber determined by plaque-assay-method after continuous injection.

Table 2. Virulence (constant injection).

Sampling time	Virulence [%] [1] of P1 (pfu/m³) [2]					
	UVC_1	UVC_2	UVC_3	CP_1	CP_2	CP_3
P1 maximum virus load	100 % ($3 * 10^5$)	100 % ($1*10^4$)	100 % ($1 * 10^6$)	100 % ($2 * 10^4$)	100 % ($1.5 * 10^6$)	100 % ($3 * 10^3$)
P2 30 min after device on	25 %	30 %	< 1 %	15 %	20 %	20 %
P3 1 h after device on	30 %	35 %	< 1 %	- [3]	50 %	< 5 %

[1] Results from fivefold determination, Rounded values: (P2)/(P1)*100; resp. (P3)/(P1)*100

[2] Rounded values of plaque forming unit's pfu related to 1 m³ sampling volume

[3] Test aborted for safety reasons

The maximum viral loads of P1 before switching on the device is given as pfu/m³ which is directly proportional to the active microbial plaques counted on the agar plates. The measured virus reductions are given as percentage values of the maximum viral load P1. The final virulence achieved by UVC technology was about 30 % without a HEPA filter. With HEPA filter almost a complete removal of the viral load was achieved. The plasma technology CP_1 was highly effective but the test had to be aborted because of high hazardous ozone concentrations. CP_2 levelled out to a final virulence of 50 %. The air cleaner CP_3 with an additional HEPA-filter reduced the final virulence to lower than 5 %.

Even with continuous aerosol release, efficient air cleaners can prevent the accumulation of infectious viral aerosol particles, thus substantially reducing the risk of infection in an entire room. Considering the general conditions of a high virus load, a reduction factor (inverse of virulence) of more than 50% can be regarded as very efficient because for every inactivated virus a fresh active virus is injected. Even higher reduction rates lead to a faster inactivation of viruses.

Analysis of by-products

In Table 3 the maximum measured ozone values and the analysed VOCs are listed. No single VOC exceeded the indoor air guidance values ("Richtwerte" RW I and RW II, AIR 2022) and all examined devices showed TVOC values lower than level 1 in accordance with the guidance value concept (UBA 2007). With except to CP_1 whose measurement was aborted, the other five investigated air cleaner can be assigned to the level 1 "hygienically acceptable".

Table 3. Increase in concentration of VOCs, TVOC and ozone.

Substance (CAS no.)	Concentrations related to device no						limit values	
	UVC_1	UVC_2	UVC_3	CP_1	CP_2	CP_3		
	[µg/m³]						**[µg/m³]**	
Ozone (10028-15-6)	0	8	2	> 600	7	4	10 [1)]	
							RW I [2)]	RW II [2)]
VOCs	**[µg/m³]**						**[mg/m³]**	
ethanol (64-17-5)	2				10		n. s.	n. s.
acetone (67-64-1)	52		50		13	5	53	160
1-propanol (71-23-8)	7						n. s.	n. s.
acetic acid (64-19-7)	9		3	[3)]			n. s.	n. s.
1-butanol (71-36-3)	11				4		0.7	2.0
ethylene glycol (107-21-1)			7				n. s.	n. s.
1,2-propanediol (57-55-6)			1				0.060	0.60
toluene (108-88-3)			1				0.30	3.0
formaldehyde (50-00-0)	3						0.10	0.10
acetaldehyde (75-07-0)	2					2	0.10	2.0
hexanal (66-25-1)					4		0.10	2.0
heptanal (111-71-7)	1					1	0.10	2.0
octanal (124-13-0)						1	0.10	2.0
TVOC [6)] (total volatile organic compounds)	87	0	62	[3)]	31	9	level 1: ≤ 0.3	

1) Maximum ozone increase according to the VDI recommendation (VDI 4300-14, 2021)

2) Indoor air guidance values ("Richtwerte" RW I and RW II, AIR, 2022), n. s. not specified

3) Test aborted for safety reasons

4) Total sum of all increased VOCs, levels according to the guidance value concept (UBA, 2007)

The ozone concentration was directly monitored by an online analyser. According to the expert recommendation (VDI-EE 4300-14, 2021), from the point of view of indoor hygiene, only a tolerable residual ozone concentration of 10 µg/m³ may be released. Almost all measured air cleaner showed lower additional ozone increase than the limit value. The released ozone concentrations of CP_1 was extremely high. Because of continuously increasing the test was aborted at round 300 ppb (> 600 µg/m³).

The technologies mentioned above are also marketed alternatively for indoor air purification of emissions. Therefore, some of the VOCs present in the room were also degraded. This was confirmed by measuring the chamber background, which was, in some cases, significantly higher than during device operation. Almost none of the investigated inactivating technologies released harmful by-products, such ozone or VOC. The measured concentrations were below the given limits. Because the technologies investigated can produce reaction products that pose a health risk, it could be assumed that the manufacturers optimized the units accordingly in advance, such as implementing suitable "ozone-free emitter". For cold plasma technologies some manufacturers installed activated carbon filters as absorbents for ozone and VOCs (Batakliev et al, 2014).

Correlation of high ozone release and formation of by-products

The air cleaner CP_1 which produced continuously increasing ozone was investigated separately because of no relating VOC and ozone values could be determined by the virulence investigations. The ionizer was adjusted in different runs to the ionization levels 100 %, 31.3 %, 25.0 % and 12.5 % of the entire ionizer power consumption of the inactivating unit. The measured ozone concentrations are displayed in Figure 1.

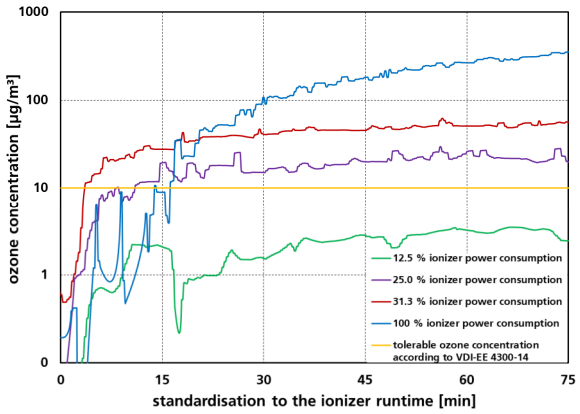

Figure 1. Course of the ozone concentrations released by CP_1.

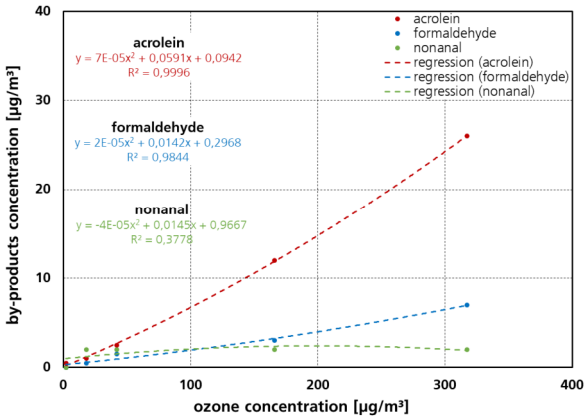

Figure 2. Correlation of ozone concentrations and formation of by-products for CP_1.

The increasing phases of the ionization took at least 5 min until the ionizer runs constantly. A low ionization power displayed in the curve of 12.5 % did not exceed the limit value of 10 µg/m³. The second level of 25 % showed a minor exceed and finally reached a constant ozone concentration at 25 µg/m³. Higher ionization levels at 31.3 % finally resulted an ozone concentration of 55 µg/m³. The highest ionizer power rate did not show a constant level within the measured time. The produced ozone concentrations increased logarithmically.

The VOC degradation was examined in a further test cycle of final 8 VOC test runs. Two background levels were adjusted prior to the operation of the device: low 272 µg/m³, high 850 µg/m³). After switching on the device two further VOC samples among ozone generation were taken. The measured ozone concentrations for the highest ionizer power consumption (100 %) were 170 µg/m³ at the second and 320 µg/m³ at the third sampling time. Independently from the ionization level, no impact of ozone on isopropanol, hexanal, pentanoic acid, and acetophenone could be seen. However a great influence to limonene was detectable. Limonene concentrations decreased about 40 % for the high background level and 50 % for the low, respectively. With the decrease in limonene concentration, an increase in the concentrations of formaldehyde and acrolein was observed. Figure 2 displays the increasing concentrations for all identified by-products directly proportional to the ozone concentrations.

The finding, that cold plasma technology can produce high concentrations of ozone and formaldehyde, was in accordance with former investigations of air cleaner (Bahri, Haghighat, 2014; Zhang et al, 2011; Guo et al, 2021). The increase of formaldehyde as product of the reaction of limonene and ozone has been studied. Its pathway is described as a complex gas phase oxidation of limonene with a huge number of intermediate products. The formation of acrolein as reaction products of limonene and ozone was not described so far, but can be explained as products as well because of different oxidized intermediate products with shorter hydrocarbon chains (Wolkoff, 2020; Carslaw, 2013).

4 CONCLUSIONS

All inactivating air cleaners on basis of UVC- or cold plasma-technology were able to reduce the viral load even among the aspect of a continuous injection of active viruses which refers to a superspreader event. Surprisingly nearly all investigated air cleaners did not release conspicuous by-products anymore. Even the additionally produced ozone concentrations were below the limit value of a tolerable residual ozone concentration of 10 µg/m³. However, one single air cleaner on cold plasma technology release ozone to a considerable extent. Depending on the high ozone concentrations the formation of the reaction products formaldehyde and acrolein could be observed in relation to the degradation of limonene. From the current state of the art, these undesired by-products could have been prevented with activated carbon filters.

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