EPA-600/R-95-154
October 1995

OZONE GENERATORS

IN INDOOR AIR SETTINGS

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Prepared for:

U. S. Environmental Protection Agency

Office of Research and Development

Washington, DC 20460

TECHNICAL REPORT DATA
(Please read Instructions on the reverse before comp

1. REPORT NO.	2.	3.
EPA-600/R-95-154		PB96-100201

4. TITLE AND SUBTITLE	5. REPORT DATE
Ozone Generators in Indoor Air Settings	October 1995
	6. PERFORMING ORGANIZATION CODE
7. AUTHOR(S)	8. PERFORMING ORGANIZATION REPORT NO.
Raymond S. Steiber	
9. PERFORMING ORGANIZATION NAME AND ADDRESS	10. PROGRAM ELEMENT NO.
See Block 12	11. CONTRACT/GRANT NO.
	NA (Inhouse)
12. SPONSORING AGENCY NAME AND ADDRESS	13. TYPE OF REPORT AND PERIOD COVERED
EPA, Office of Research and Development Air Pollution Prevention and Control Division Research Triangle Park, NC 27711	Final; 6/92 - 12/93
	14. SPONSORING AGENCY CODE
	EPA/600/13

15. SUPPLEMENTARY NOTES Author Steiber's mail drop is 54; his phone number is 919/541-2288.

16. ABSTRACT The report gives information on home/office ozone generators. It discusses their current uses as amelioratives for environmental tobacco smoke, biocontaminants, volatile organic compounds, and odors, and details the advantages and disadvantages of each. Ozone appears to work well against household odors and environmental tobacco smoke, but caution needs to be exercised in its use because of the production of byproducts such as formaldehyde. Ozone has biocidal effects, but its use in household settings is limited by the high concentrations needed for complete kills. Ozone has decremental effects on lung function in humans that persist for 24-28 hours. In the experiments conducted at the indoor air test house, each of the three ozone generators studied produced concentrations in excess of the Occupational Safety and Health Administration limit for workplace exposures. In addition, when interior doors were left open, adjoining rooms were also subjected to such exposures. Total ozone decay times for all the concentrations studied did not exceed 12 minutes.

17. KEY WORDS AND DOCUMENT ANALYSIS

a. DESCRIPTORS		b. IDENTIFIERS/OPEN ENDED TERMS	c. COSATI Field/Group	
Pollution	Volatility	Pollution Control	13B	20M
Ozone	Organic Compounds	Stationary Sources	07B	07C
Generators	Odors	Ozone Generators	14G	06P
Tobacco		Biocontaminants	06C, 02D	
Smoke	Biocides		21B	21D
Contaminants				

18. DISTRIBUTION STATEMENT	19. SECURITY CLASS (This Report)	21. NO. OF PAGES
Release to Public	Unclassified	34
	20. SECURITY CLASS (This page)	22. PRICE
	Unclassified	

EPA Form 2220-1 (9-73)

FOREWORD

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The National Risk Management Research Laboratory is the Agency's center for investigation of technological and management approaches for reducing risks from threats to human health and the environment. The focus of the Laboratory's research program is on methods for the prevention and control of pollution to air, land, water, and subsurface resources; protection of water quality in public water systems; remediation of contaminated sites and groundwater; and prevention and control of indoor air pollution. The goal of this research effort is to catalyze development and implementation of innovative, cost-effective environmental technologies; develop scientific and engineering information needed by EPA to support regulatory and policy decisions; and provide technical support and information transfer to ensure effective implementation of environmental regulations and strategies.

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E. Timothy Oppelt, Director
National Risk Management Research Laboratory

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SUMMARY

This report presents information on home/office ozone generators. It discusses their current uses as amelioratives for environmental tobacco smoke (ETS), biocontaminants, volatile organic compounds, and odors and details the advantages and disadvantages of each. Ozone appears to work well against household odors and ETS, but caution needs to be exercised in its use because of the production of byproducts such as formaldehyde. Ozone has biocidal effects, but its use in household settings is limited by the high concentrations needed for complete kills. Ozone has decremental effects on lung function in humans that persist for 24-48 hours.

In experiments conducted at the indoor air test house, each of the three ozone generators studied produced concentrations in excess of the Occupational Safety and Health Administration limit for workplace exposures. In addition, when interior doors were left open, adjoining rooms were also subjected to such exposures. Total ozone decay times for all the concentrations studied did not exceed 12 minutes.

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CHAPTER 1

OZONE AS AN INDOOR AIR AMELIORATIVE

Ozone generators are advertized by their manufacturers as cure-alls for a large number of indoor air complaints. They are claimed to be effective at eliminating environmental tobacco smoke (ETS), biocontaminants such as molds and bacteria, volatile organic compounds (VOCs), and odors. Such claims are generally accompanied by customer testimonials that describe the results of using the generators in glowing terms. These generators can be found in a wide variety of settings. Hotels and motels use them to get rid of the odor of stale tobacco smoke and thus provide their guests with a "smoke-free" environment. Some hotels even mount them permanently in their rooms, usually hidden behind a grill near the ceiling. Although individual practices undoubtedly vary, many of these may be in continuous operation at low settings.

Other venues in which ozone generators are used include restaurants, offices, bars, and homes. They are used in schools to deal with molds and bacteria and in nursing homes to eliminate the odors associated with the illnesses of the aged. And yet, despite this widespread use and the many claims of effectiveness, there is little or no supporting data in the scientific literature. This does not mean that the claims are untrue, but merely that they are as yet unsubstantiated.

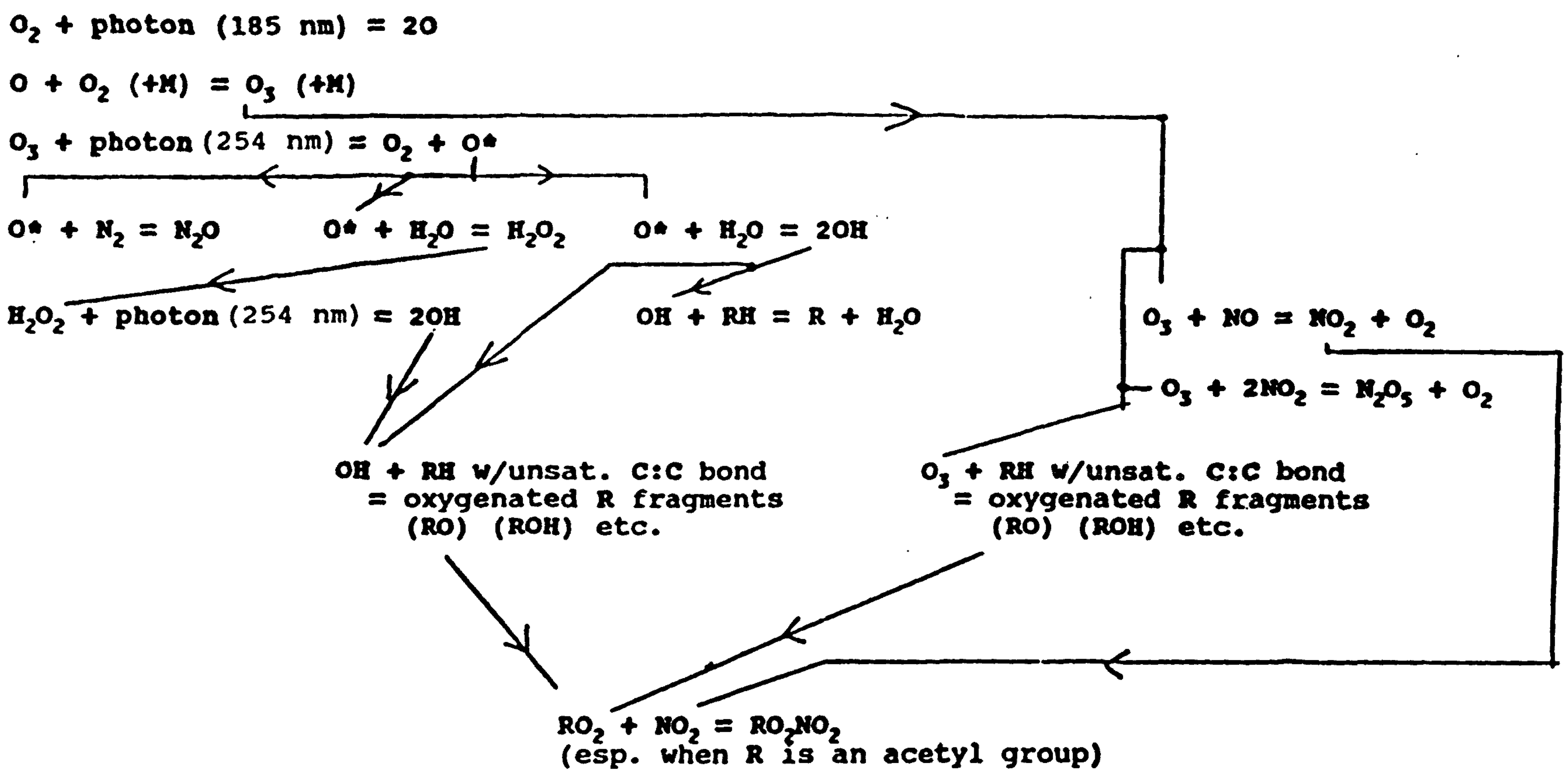
OZONE CHEMISTRY

Ozone (O_3) is produced by either the electrical or ultraviolet (UV) irradiation of the normal oxygen molecule (O_2). If nitrogen is present, the electrical discharge method will co-produce nitrogen oxides. O_3 is a more active oxidant than O_2, but this does not mean that it readily oxidizes every class of organic compound exposed to it (alkanes, for instance, do not react with ozone at all) or that the products of such oxidations will be innocuous. Ozone reacts rapidly with alkenes (olefins) to form aldehydes and ketones. It also reacts rapidly with alkynes to produce carboxylic acids. It reacts with water (H_2O) molecules to form hydrogen peroxide (H_2O_2) and with nitrogen oxide (NO) and nitrogen dioxide (NO_2) to form NO_2 and nitrogen pentoxide (N_2O_5), respectively. The process by which ozone reacts with organic compounds is known as ozonolysis. In the case of alkenes and alkynes, ozone reacts at the unsaturated carbon:carbon bonds, cleaving the molecule and forming separate oxygenated byproducts. Except at extreme concentrations where a continuous chain of reactions might eventually exhaust the available organic material, leaving behind carbon dioxide (CO_2) and water, it is not true that ozonation destroys volatile organic compounds as a class. Rather it reacts with them to form new groups of VOCs, some of which may actually be more toxic[1] than the compounds they have replaced. This statement is substantiated by studies[2] of polluted airsheds in which ozone plays an important role in the formation of the compounds that constitute smog.

Figure 1 shows a typical UV-initiated ozone reaction chain. The reactions shown are based on reports[3,4] in the literature and do not, by any means, constitute the only ones that can occur. In this case, the chain is started when a normal O_2 molecule is cleaved due to irradiation by ultraviolet light in the 185 nanometer (nm) band. The single oxygen atoms join with other O_2 molecules to form O_3. Further irradiation in the 254 nanometer band results in the formation of a highly reactive oxygen singlet (O*). This singlet is important in that it reacts with H_2O to produce both hydrogen peroxide and hydroxyl ($OH^{\cdot}$) radicals, both of which play a role in further reactions (the reaction that produces N_2O is speculation on the part of the author and may not occur in nature). As can be seen, UV-initiated ozone reactions are complex and set the stage for later reactions that, far from destroying VOCs, may result in the formation of more complex organics, some of which may be nitrated.

OZONE AND FORMALDEHYDE

Formaldehyde is a toxic agent with a Threshold Limit Value (TLV) of 1 part per million in air. It is also suspected of being a carcinogen. Formaldehyde is one of a number of aldehydes that can be formed when ozone reacts at the unsaturated carbon:carbon



...and so forth

O* : highly reactive, short-lived oxygen singlet

FIGURE 1. Ultraviolet-initiated ozone reaction chain.

bonds of an olefin. Olefins constitute a significant portion of the hydrocarbons found in mineral oil, and mineral oil is widely used as a solvent for binders in home furnishings and building materials. As a consequence, olefins are ubiquitous in the VOC background of any office or household. Therefore, it seems likely that the presence of any amount of ozone within a dwelling, whether it comes from outside air, electrical appliances, or ozone generators, will result in the formation of some concentration of aldehydes.

In a study conducted by Weschler et al.[5], carpet was exposed to parts per billion concentrations of ozone in a stainless steel chamber. The concentrations of formaldehyde, acetaldehyde, and aldehydes with higher carbon numbers increased significantly in the chamber headspace. At the same time there was a corresponding decrease in unsaturated VOC concentrations. In a follow-up study conducted by Zhang and Lioy[6] in six residential houses, formation of the same series of aldehydes was observed in the presence of ozone. However, increases in formaldehyde concentrations were relatively insignificant when compared to those emitted by other sources in the households (formaldehyde is used in fabrics, particle board, plywood, and other products). Zhang and Lioy also observed increases in the presence of formic acid, particularly as indoor humidity increased.

ODOR CONTROL

Ozone is documented as being effective in eliminating unpleasant tastes and odors in drinking water. Persson et al.[7] have shown that, at concentrations of 4.0 milligrams/liter, ozone will remove 80-90% of the geosmin and 2-methyl isoborneol (MIB) found in the water. These bacterially produced alcohols impart particularly strong tastes and odors. Ozone is also an effective oxidant for reduced sulfur compounds, another bad tasting and bad smelling ingredient of some drinking waters. In addition, ozone works well against reduced chlorine compounds such as trihalomethanes.

MIB, geosmin, and reduced sulfur compounds such as dimethyl disulfide are found in indoor air and account for many of the household odors associated with biocontamination. They are most frequently associated with actinomyces, such as streptomyces sp., a fungus-like bacterium that is also found in soils. In water, contact time for the elimination of these chemicals by ozone is 6-12 minutes, but destruction in household air would probably take much longer due to greater dilution in the vapor state.

Ozone is claimed to be effective in combatting the odors of tobacco smoke and is widely used for that purpose with apparent success. Researchers[8] have identified large numbers of separate compounds as components of ETS. These include nitrosamine, polynuclear aromatic hydrocarbons, nicotine, phenols, ketones, and a host of others. In terms of odor, however, the aldehyde acrolein, C_3H_4O, is probably the most important. Acrolein has a pungent odor and causes eye and nose irritation. It is acrolein that leaves a raw, burning flavor in the mouths of heavy smokers. Acrolein is not very stable, and, although no supporting data presently exist, one can speculate that it readily reacts with ozone. If this is indeed the case, it would go far in explaining ozone's claimed effectiveness as an ameliorative for ETS.

Users of ozone generators frequently comment on the pleasant odor they leave behind after they have been turned off. Since ozone rapidly decays to the normal oxygen molecule (10-12 minutes for most concentrations), this odor is not the ozone itself but some combination of its most common reaction products. The odor is frequently described as a "clean sheet" smell or the odor that clothing gives off when it is fresh out of the drier. The exact nature of the compounds that cause this odor is not known, but one can speculate that they may be some combination of hydrogen peroxide and the oxides of nitrogen. At low concentrations some of the oxides of nitrogen, particularly nitrous oxide, have a pleasant, sweetish smell. In addition, they are known to cause mild feelings of euphoria.

OZONE AS A BIOCIDE

Ozone has been used since 1903 in France and since 1934 in the U.K. as a biocide in the purification of drinking water. In recent years, ozone has also been touted as a biocide for molds and bacteria in the household. The typical ozone generator marketed for home use develops concentrations of from 0.1 to 0.4 part per million in the average room. These numbers assume an air exchange rate with the outside of 0.25 to 0.5 room air exchanges per hour. Somewhat larger generators are used by commercial duct cleaners, but even here concentrations seldom exceed 1 to 2 parts per million. (Some especially large units that are mainly used for eliminating the odors left behind by building fires are claimed to operate in the 20 parts per million range.)

Foarde et al.[9] conducted chamber experiments in which the spores of two separate fungi (Penicillium chrysogenum and Penicillium glabrum) were exposed to selected concentrations of ozone for 24 hours at two relative humidities (RHs), 30% and 90%. In addition, the spores of the spore-forming bacterium Streptomyces sp. and a living yeast, Rhodotorula glutinis, were exposed in the same manner. Destruction at such a level as to preclude either regrowth or survival did not begin to occur until ozone concentrations had reached the 5-10 parts per million range. The kill ratios were very much RH dependent, with greater concentrations of ozone required at the lower RH. When ceiling tiles were used as a substrate for the deposition of the spores, kill ratios dropped even lower. Ozone, then, does have biocidal effects, but for it to be used successfully in household settings, much higher concentrations must be applied than are now generally the case. Given the collateral effects, such as damage to rubber products and the formation of byproducts, it remains to be seen whether such concentrations are practical.

HEALTH EFFECTS

In medium to high concentrations (0.5 to >1 ppm) ozone is known to cause irritation to the eyes and the mucous membranes. This is probably due to its desiccating effects. In its criteria document[10] for ozone and other photochemical oxidants the Environmental Criteria and Assesssment Office (ECAO) of the U.S. Environmental Protection Agency (EPA) summarizes the results of a large number of studies involving ozone exposures of humans and animals. These studies paid particular attention to reductions in lung function. In general, they reported decremental effects in adults at exposures of 0.37 part per million ozone for 1-3 hours. A 50% recovery of function took place within a few hours of the exposures' having ended and a full recovery within 24 hours. Repeated exposures for long periods of time (6 months to a year) prolonged full recovery times up to 48 hours. Similar responses were noted in children and the elderly at concentrations as low as 0.14 part per million. Most of these studies involved some period of exercise during the exposure cycle, usually 15 minutes of exercise followed by 15 minutes of rest.

The EPA ambient clean air standard for ozone is 0.12 part per million for 1 hour. The World Health Organization's air quality guidelines for Europe set a limit of 0.0 75 to 0.1 part per million for 1 hour. The Occupational Safety and Health Adminstration sets the workplace ozone exposure limit at 0.1 part per million for 8 hours.

SUMMARY

Ozone is known to be useful as a biocide for drinking water and wastewater. However, its use as a biocide in indoor air settings is limited by the high concentrations that are needed. Ozone appears to work well against household odors caused by biocontaminants and ETS; but, because of its byproducts (formaldehyde, nitrogen oxides etc.), caution needs to be exercised in its use (at the very least any room so exposed should be aired afterwards, and such exposures should not take place while the room is occupied). Ozone has decremental effects on lung function, but once the exposure is ended there is a full recovery within 24-48 hours. In outside air, ozone promotes many of the reactions that result in smog. Some of these reactions must also occur in indoor air, and further studies are needed in this area.

CHAPTER 2

TEST HOUSE STUDIES OF OZONE GENERATORS

The Indoor Air Branch (IAB) of the National Risk Management Research Laboratory's Air Pollution Prevention and Control Division maintains a test house where indoor air studies are carried out. The house is a three-bedroom, single-storey frame dwelling located in a typical East Coast suburb. It has an attached garage and a crawl space instead of a basement. Figure 2 shows the floor plan.

This chapter describes a series of experiments carried out in the test house using commercially available home/office ozone generators. The purpose of these tests was to document outputs and transport in a household setting, and no other types of measurements were taken. IAB did not, for instance, attempt to ascertain what chemical reactions might be taking place.

OUTPUTS OF THREE OZONE GENERATORS

Three models of home/office ozone generators were tested under the same general conditions. For the purposes of this report they will be identified as Unit A, Unit B, and Unit C. Each is equipped

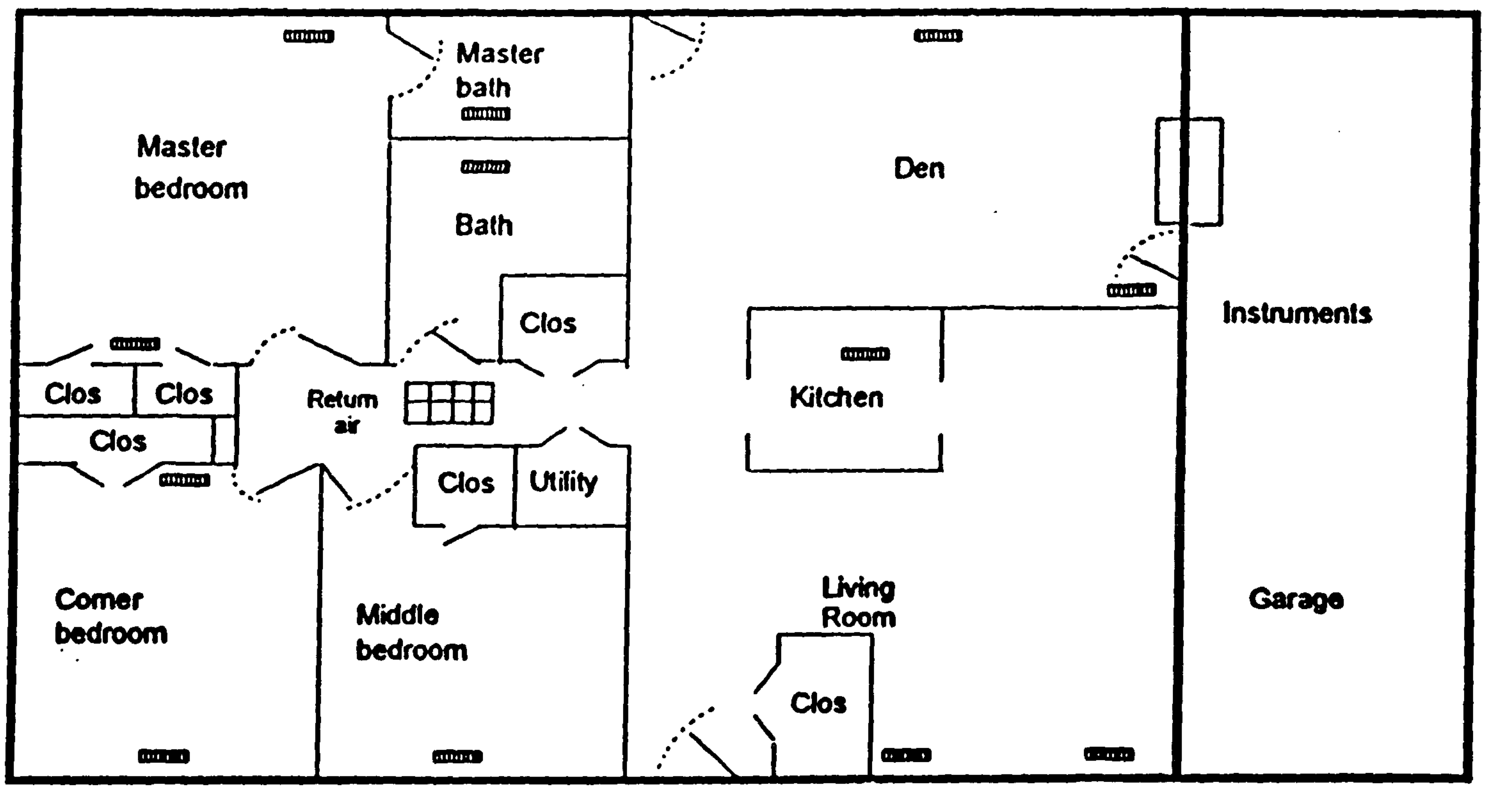


FIGURE 2. Test house floor plan.

with controls that allow the user to select fan speeds and ozone output. In addition, Unit C comes equipped with three insertable generator plates that allow the user to alter the output range of the unit. In order to simplify the text, these plates will be designated Configurations 1, 2, and 3.

Before testing began, a longitudinal and latitudinal traverse was made of the faces of each of the units at the low, medium, and high settings. This was done using a TECO Model 565 Ozone Analyzer. The measurements were averaged and the output calculated in terms of milligrams per hour. The fan speeds were set at medium, and the average air speeds were determined with an Alnor Model 8565 hot wire anemometer. Table 1 presents the results.

TABLE 1. Ozone Generator Outputs at the Face in Milligrams per Hour.

UNIT	CONFIGURATION	LOW	MEDIUM	HIGH
A	N/A	0.00696	0.02690	0.06720
B	N/A	0.01992	0.11460	0.74370
C	1	BLD*	0.00148	0.03140
C	2	0.00443	0.02772	0.29298
C	3	0.00554	0.06282	0.57264

*below limit of detection

As can be seen, Unit C: Configuration 1: Low Setting had the least output while Unit B: High Setting had the greatest. It is interesting to note that for Units B and C the difference in outputs between the medium and high settings is not double, as one would expect, but 6 to 10 times greater.

The first set of experiments at the test house were run for the purpose of determining the maximum sustainable ozone concentrations that could be achieved in a closed room by each unit in each of its settings and configurations. The room selected for the tests was the front corner bedroom (see Figure 2). This room has a volume of slightly more than 27 cubic meters. All doors and windows remained shut during the tests, and the heating, ventilating, and air conditioning (HVAC) system was off. Air exchange rates were determined using the tracer gas decay method and averaged 0.3 air exchanges per hour. Each test was run in duplicate and had a duration of at least 90 minutes. Output averaging did not begin until concentrations in the room had reached equilibrium. This normally took 15-20 minutes. The generator fan speeds were set at medium.

Table 2 shows the results for Unit A. All the data in this and subsequent tables have, of course, been corrected for the background.

TABLE 2. Unit A: Maximum Sustainable Ozone Concentrations in Parts per Billion.

TEST	OUTPUT SETTING	AVERAGE OUTPUT	HIGHEST SPIKE
1	low	8	40
2	low	14	60
3	medium	40	64
4	medium	28	54
5	high	200	320
6	high	180	480

Table 3 shows the same type of data for Unit B.

TABLE 3. Unit B: Maximum Sustainable Ozone Concentrations in Parts per Billion.

TEST	OUTPUT SETTING	AVERAGE OUTPUT	HIGHEST SPIKE
7	low	4.5	22
8	low	6.6	21
9	medium	22	46
10	medium	15	36
11	high	222	420
12	high	204	390

Table 4 shows the results for all three of the configurations of Unit C.

TABLE 4. Unit C: Maximum Sustainable Ozone Concentrations in Parts per Billion.

TEST	SETTING	CONFIGURATION	AVERAGE OUTPUT	HIGHEST SPIKE
13	low	1	BLD*	--
14	low	1	BLD*	--
15	medium	1	2.2	4.6
16	medium	1	1.8	5.8
17	high	1	19	32
18	high	1	16	24
19	low	2	2	6
20	low	2	2	3
21	medium	2	10	15
22	medium	2	10	20
23	high	2	140	200
24	high	2	180	240
25	low	3	8	18
26	low	3	8	28
27	medium	3	20	35
28	medium	3	15	46
29	high	3	140	200
30	high	3	290	460

*below limit of detection

Test 29 appears to be an outlier. Configuration 3 requires the insertion of two plates in the unit, and if one of them were not making proper contact, the unit would operate as if it was set up in Configuration 2.

OZONE TRANSPORT

A series of experiments were run to determine how ozone concentrations in one area of the test house would add to ozone concentrations in another area. Since ozone decays to O_2, this cannot easily be calculated using interior and exterior air exchange rates and mass balance equations.

In the first group of experiments, Unit C: Configuration 3 was set up on a counter in the kitchen of the test house (see Figure 2). An ozone detector was placed in the front corner bedroom. Tests were then run under four conditions:

Bedroom door open, HVAC off
Bedroom door closed, HVAC off
Bedroom door open, HVAC on
Bedroom door closed, HVAC on

The ozone generator controls were set at maximum ozone output and maximum fan speed, and all exterior doors and windows were shut. Table 5 presents the results.

TABLE 5. Transport Tests, Group 1: Ozone Concentrations in Parts per Billion.

CONDITIONS	AVERAGE CONCENTRATION	HIGHEST READING	LOWEST READING
bedroom door open, HVAC off	50	72	18
bedroom door closed, HVAC off	2	4	0
bedroom door open, HVAC on	28	38	14
bedroom door closed, HVAC on	38	54	26

In the tests run with the bedroom door open and the HVAC off, there was a constant cycling between the highest and the lowest readings. All circulating fans have high-low cycles. This is probably due to the turbulence they create. In this case the cycling appears to have been accentuated by the distance between the generator and the detector (approximately 8.5 meters). When the HVAC system was turned on, however, a considerable smoothing took place. This may have been caused by the ductwork acting as a laminar flow element. It is also possible that the HVAC fan cycle and the generator fan cycle cancelled each other out.

In another group of transport experiments, Unit B was set up in the corner front bedroom and allowed to achieve equilibrium concentrations. Measurements were then taken in the following locations:

Master Bedroom

Middle Bedroom

Hall

Den

Living Room

Tests averaged 2-3 hours and a minimum of six readings were taken in each location. These were so consistent that, in the tables that follow, only one reading for each test will be presented. In each of these tests all the interior room doors were open and all the windows and exterior doors were shut. The HVAC system was off in the first group of tests and on in the second. All tests were run in duplicate. Tables 6 and 7 present the data.

TABLE 6. Ozone Transport, HVAC Off: Concentration in Parts per Billion.

LOCATION	**CONCENTRATION (fan max, gen max)**	**CONCENTRATION (fan med, gen max)**
Master Bedroom	120	180
Middle Bedroom	62	70
Hall	120	190
Den	32	16
Living Room	25	26

In Table 6 the smaller concentrations seen with the generator fan set at maximum are probably due to air dilution. In Table 7 below an even greater dilution is seen with the HVAC fan in operation. Concentrations, however, are more consistent due to better distribution throughout the house.

TABLE 7. Ozone Transport, HVAC On: Concentration in Parts per Billion.

LOCATION	CONCENTRATION (fan max, gen max)	CONCENTRATION (fan med, gen max)
Master Bedroom	42	no tests run
Middle Bedroom	25	"
Hall	22	"
Den	24	"
Living Room	21	"

OZONE DECAY

The ozone molecule is unstable and has a relatively short half life. Unless constantly replenished, ozone concentrations will quickly diminish to background levels. This makes ozone attractive as an ameliorating agent since, unlike other forms of treatment, there is no residue of the agent itself (however, as discussed in Chapter 1, there are reaction products).

This section presents data on ozone decay that were generated as a byproduct of the previously described tests. The decay rates presented in the graphs below are a product of both ozone's natural decay rate and the air exchange rate in the test house during the test period (0.3 air exchange per hour). In this respect they present useful data on the decay rates that can be expected in household settings. All the data were taken with all exterior doors and windows closed.

Figure 3 shows two representative decay curves from more than 30 generated. These particular curves are for ozone concentrations in the 160-200 parts per billion range. Notice that the curves are not asymptotic. This indicates that leakage (air exchange) is not the dominant process taking place. The variations at the lowest concentrations may be due to fluctuations in background.

Figure 4 presents the same sort of curves for concentrations in the 20-30 parts per billion range. Note that it takes nearly 2 minutes longer for concentrations to reach zero. This is because, at the lower measurement range, the instrument detects concentrations that at the higher ranges would appear to be zero. As can be seen, the decay of non-replenished, parts per billion concentrations of ozone in a household setting is rapid and complete.

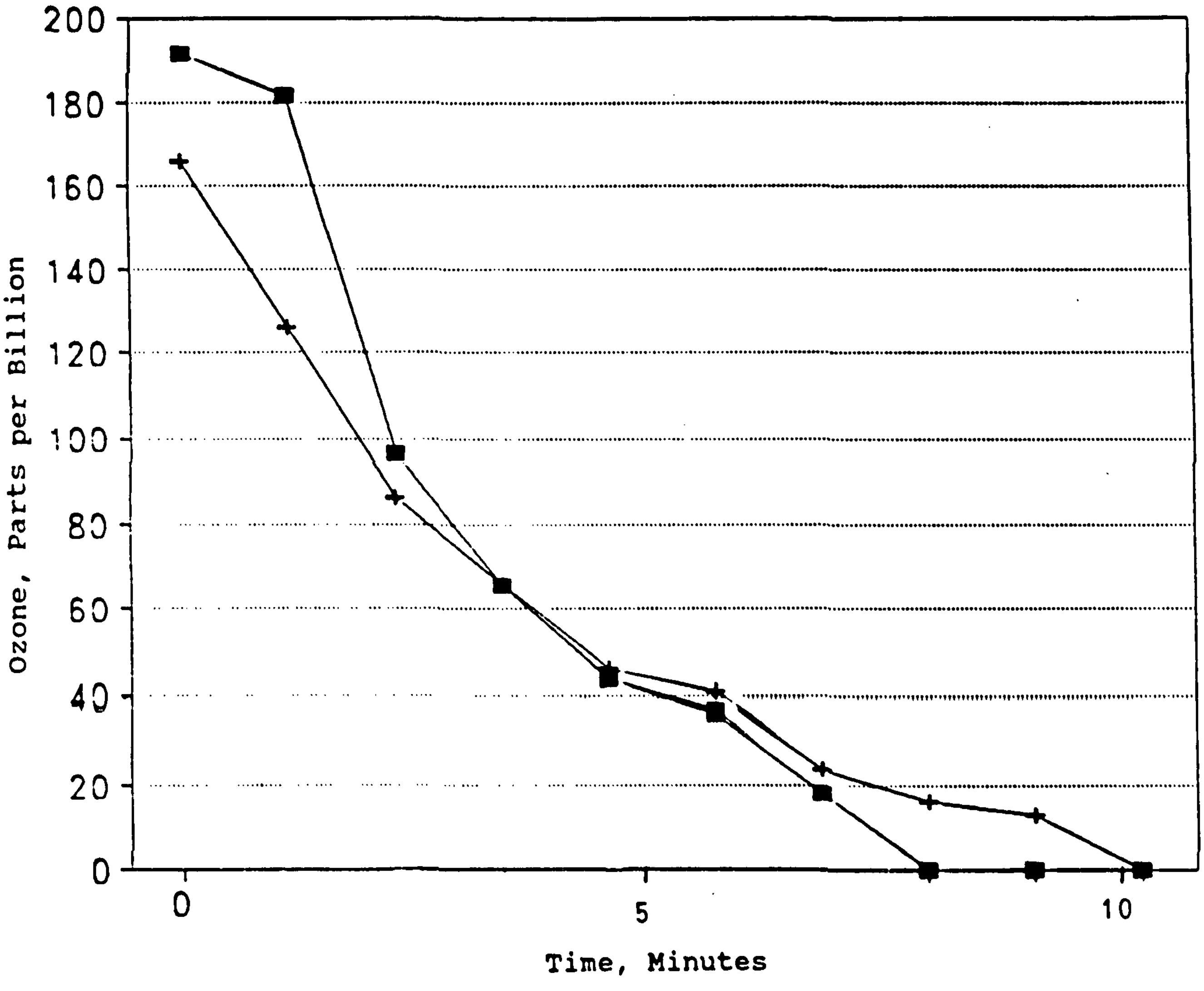


FIGURE 3. Ozone decay curves: high range.

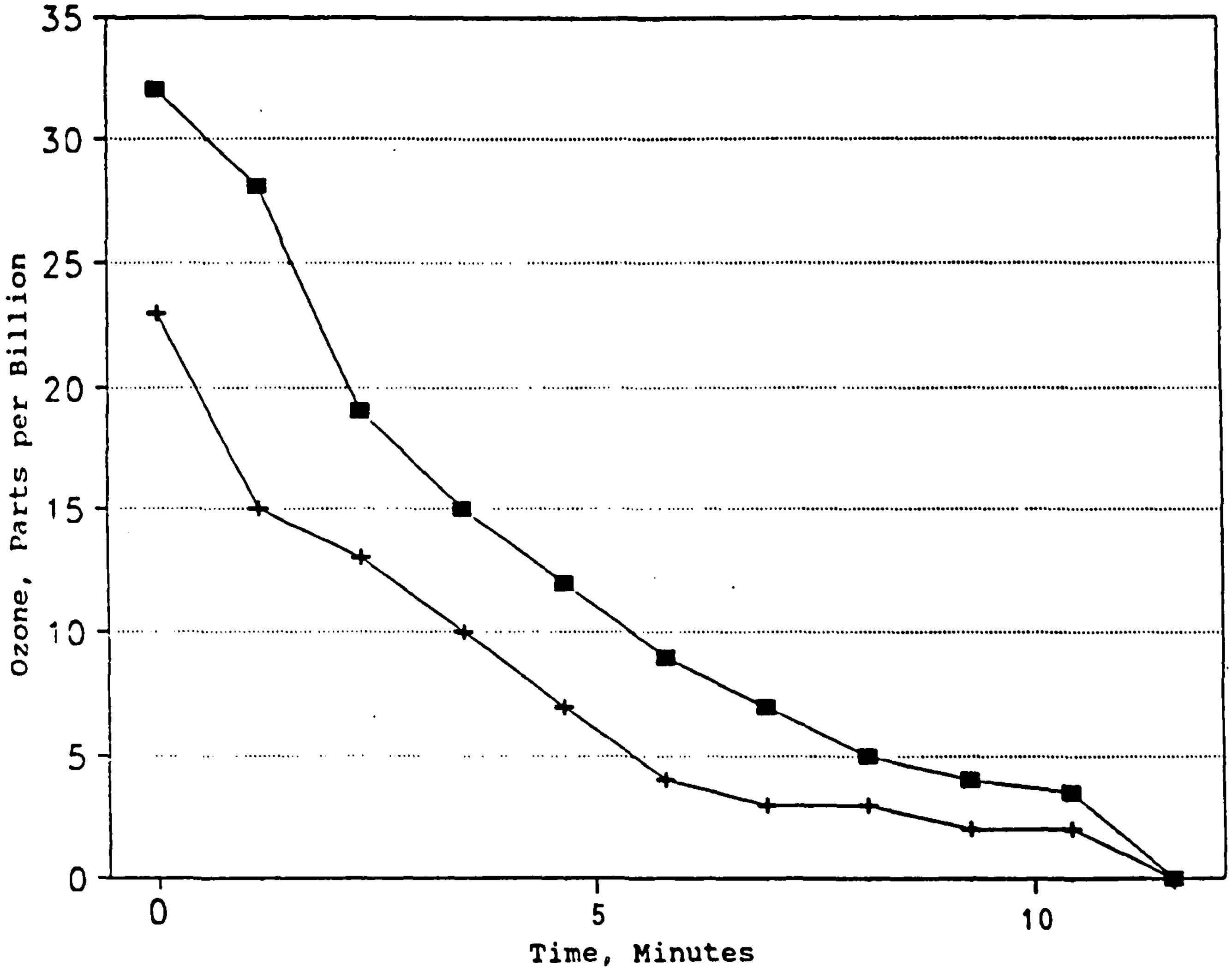


FIGURE 4. Ozone decay curves: low range.

UMMARY

Assuming a continuous 8-hour exposure in a single room etting, each of the three ozone generators exceeded the OSHA orkplace exposure limit of 0.1 part per million (100 parts per illion) by 1.5 to 2 times at their maximum settings. The Food and rug Administration sets an exposure limit for medical devices, ncluding air cleaners, of 0.05 part per million (50 parts per illion). At their maximum settings the units exceeded this limit y nearly 4 times.

In addition, when interior house doors were left open, djoining bedrooms and the hall were exposed to concentrations that xceeded one or both of these limits.

For all the concentrations examined, total ozone decay times id not exceed 12 minutes.

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APPENDIX

Quality Assurance Statement

The work reported in Chapter 2 was covered by the quality assurance plan for test house gas measurements that was in effect in the period 1991-1992, and no separate quality assurance plan was submitted.

The following practises were followed to ensure data quality. The ozone detector, a TECO Model 560 Ozone Analyzer, was calibrated against an ozone photometer in accordance with 40 CFR, Part 50, Appendix D. The photometer that was used is one that is periodically checked against the National Institute of Standards and Technology's standard photometer. Variations between the two instruments did not exceed 0.001 ppm at any concentration. The ozone analyzer zero was checked at the beginning and end of each day's sampling and whenever the range was changed. Background ozone levels were determined at the beginning and end of each test, averaged, and subtracted from the total measured. Only those data taken after room ozone concentrations had reached equilibrium (10-15 minutes) are reported in the chapter. Duplicate, and in some cases triplicate, tests were run under each set of conditions. Data from single room tests and some multi-room tests were recorded on a strip chart recorder and the strip charts preserved. Data from other multi-room tests were taken manually and recorded in the sampling log book. Information on times, weather, backgrounds, sampling conditions, and a number of other factors was recorded in the sampling log book and the book preserved. Data on air exchange rates are a part of the permanent test house data base and are recorded on disk.

The configuration of the single room tests was as follows. The ozone detector was placed at the center of the front wall of the room with the sample intake tube at a height of approximately 3.5 feet. The ozone generator was situated approximately 2 feet from the center of the back wall on a stand approximately 4.5 feet high. Previous tests with tracer gases run over several years had determined that this was a well-mixed room and that, once equilibrium had been reached, there would be no pockets of higher or lower concentrations. In the multi-room tests the ozone generator was placed in the center of the room and faced the door.