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Summary: *Acceptable indoor air quality in office buildings may be achieved with less energy by combining effective air cleaning systems for volatile organic compounds (VOCs) with particle filtration then by relying solely on ventilation. For such applications, ultraviolet photocatalytic oxidation (UVPCO) systems are being developed for VOC destruction. An experimental evaluation of a UVPCO system is reported. The evaluation was unique in that it employed complex mixtures of VOCs commonly found in office buildings at realistically low concentrations. VOC conversion efficiencies varied over a broad range, usually exceeded 20%, and were as high as ~80%. Conversion efficiency generally diminished with increased air flow rate. Significant amounts of formaldehyde and acetaldehyde were produced due to incomplete mineralization. The results indicate that formaldehyde and acetaldehyde production rates may need to be reduced before such UVPCO systems can be deployed safely in occupied buildings.*

Keywords: *air cleaning, clean air delivery, formaldehyde, office buildings, volatile organic compounds*
Category: *technologies for healthy buildings*

1. Introduction

In many types of commercial buildings, the indoor generated air pollutants of concern are primarily volatile organic compounds (VOCs) and various types of particles. Implementation of practical air cleaning technologies for both VOCs and particles may improve indoor air quality (IAQ) or enable IAQ levels to be maintained with reduced outdoor air supply and concomitant energy savings. Unlike ventilation, air cleaning also can reduce indoor exposures to outdoor pollutants.

Practical air cleaning technologies for particles are available, typically consisting of fibrous filters installed in incoming outdoor air and recirculated air streams. Except in the mildest of climates, the cost and energy use of efficient particle filtration is much smaller than the cost and energy use of an equivalent amount of ventilation. Consider a typical US office building with a ventilation rate of 1 h^{-1} and an air recirculation rate of 3 h^{-1}. If the ventilation rate is reduced to 0.5 h^{-1}, one needs to increase the particle removal efficiency of filters in the recirculated air by 17% to prevent an increase in concentrations of indoor-generated particles. The cost of filtration necessary to supply the equivalent of 1 m^3 h^{-1} of particle-free air (considering the most difficult to remove 0.2-μm diameter particles) is about $0.07 per person per year [1]. In comparison, the annual energy cost of 1 m^3 h^{-1} of ventilation in US office buildings is about $0.28 to $0.45 per person [1].

Currently, the effectiveness and practicality of air cleaning technologies for VOCs are not well established. One potential VOC air cleaning technology is ultraviolet photocatalytic air cleaning (UVPCO). In basic terms, UVPCO often utilizes a honeycomb configured monolith reactor coated with titanium dioxide (TiO_2 or titania) as the photooxidative catalyst. The coated monolith is irradiated with UV light from fluorescent bulbs operating near either 254 nm (UVC) or 365 nm (UVA). Air flows through the monolith, where the VOCs adsorb on the catalyst. The UV light, interacting with the catalyst in the presence of oxygen and water vapor, produces hydroxyl radicals. Hydroxyl radicals are highly chemically reactive and, in-turn, breakdown the adsorbed VOCs, ideally producing only carbon dioxide and water as products.

To be practical, a UVPCO system must effectively remove indoor generated VOCs of concern at comparable or lower cost than ventilation. The required VOC removal efficiency is the same as discussed for particle filtration. The UVPCO system also must not produce significant amounts of harmful VOCs through incomplete decomposition.

UVPCO technology has been studied almost exclusively in laboratory settings. The large majority of these investigations have employed relatively high concentrations of only few VOCs, often in an attempt to better understand the photocatalytic process. Comprehensive evaluations of the performance of UVPCO devices, under real or simulated-real indoor conditions where many VOCs are present at low concentrations generally have not been reported in the open literature. This paper describes research performed to help fill this knowledge gap.

2. Methods

The experimental methods are described in detail elsewhere [1]. The UVPCO reactor is a prototype developed to demonstrate air purification capabilities when installed in a HVAC duct system. Degussa Titania P25 impregnated with 3% tungsten oxide by weight (TiO2 / 3% WO3) is used as the photocatalyst. The device contains two aluminum honeycomb monoliths with 64 cells per square inch. The monoliths are wash coated with the photocatalyst. Each monolith has face dimensions of 30 by 30 cm and is 2.5 cm thick. The monoliths are mounted in series with their faces oriented transversely to the air flow path. Newly prepared monoliths were installed and were used throughout the first seven experiments and then changed. Air flow straightener elements are placed upstream and downstream of the monoliths.

Twelve UVC lamps are used. These are 18-Watt lamps with about 30% efficiency. Total UV power is about 5 – 5.5 Watts/lamp, predominantly at 254 nm. Device power consumption with the lamps on is 220 watts. The lamps are mounted transversely in three banks of four lamps each. The banks are centered between the monoliths as well as before the first and after the last monolith. The distance between a lamp surface and the face of a monolith is ~2.5 cm. This arrangement results in reasonably uniform light intensity distribution over the monolith faces. Since photocatalysis depends on approximately the square root of intensity, no significant performance differences accrue due to the small differences in light intensity over the various monolith cells. The lamps had approximately 1,000 h of prior use, so no significant change in lamp intensity was anticipated over the study. A pleated fabric air filter (MERV 12 rating) is installed at the air inlet.

The UVPCO device was separately challenged with three mixtures of VOCs typical of mixtures encountered in indoor air. A synthetic office mixture contained 27 VOCs commonly measured in office buildings [2]. A cleaning product mixture contained three common cleaning products. These were a pine-oil based cleaner, a cleaner utilizing 2-butoxyethanol (2-BE) as the solvent, and an orange-oil (i.e., d-limonene) based cleaner combined in a ratio of 2:1:1, respectively. A mixture of building product VOCs was created by combining sources including painted wallboard, composite woods, carpet systems, and vinyl flooring in a ventilated chamber. A total of nine experiments were conducted using the three mixtures. Two other experiments were conducted with just formaldehyde and acetaldehyde in inlet air. With steady-state VOC concentrations, air was drawn through the UVPCO device and single pass conversion efficiencies were measured. Air flow rates and concentrations of the mixtures were manipulated, with concentrations of individual VOCs mostly maintained below 10 ppb. Inlet air relative humidity (RH) ranged from 42% to 65% and inlet temperature was 19.5 to 25 ^{o}C.

Air flow rate, pressure drop across the system (monoliths, lamps and straighteners) temperature, and RH were continuously monitored. Simultaneous air samples for the analysis of VOCs, low molecular weight carbonyls, and low molecular weight carboxylic acids were collected in triplicate both upstream and downstream of the reactor section. VOC air samples were collected onto tubes packed with Tenax-TA™ with a section of Carbosieve™ S-III at the outlet. Carbonyl samples were collected onto silica gel cartridges treated with 2,4-dinitro-phenyhydrazine. Silica gel cartridges treated with sodium hydroxide were used for sampling acids. All sampling air flow rates were regulated with electronic mass flow controllers. VOC samples were analyzed by thermal desorption gas chromatography with mass selective detection and quantitation (TD-GC/MS) [3]. Most analytes were quantified using calibration curves developed from pure compounds. Carbonyl samples were extracted and analyzed for formaldehyde, acetaldehyde and 2-propanone (acetone) [4]. Formic and acetic acids were extracted with water and analyzed by ion chromatography [5].

Single-pass conversion efficiency, which represents the fraction of a compound removed from the air flowing through the reactor (i.e., the fraction reacted), was calculated for each analyte. This value was determined as one minus the quotient of the average outlet concentration and the average inlet concentration. The standard deviations of all calculated quantities were determined by error propagation. A clean air delivery rate (CADR) in m^3/h was computed from the single-pass conversion efficiency as the fraction of a compound reacted multiplied by the air flow rate through the device in m^3/h.

3. Results

Experiments 1, 5, 6 and 7 were conducted with the synthetic office VOC mixture. The concentration of the VOC mixture and the air flow rate through the UVPCO reactor were varied across the four experiments. The inlet VOC mixing ratios (ppb) and the fractions of each compound reacted are shown for in Table 1. Each value is the mean plus or minus one standard deviation of the measurement. The fraction reacted is not shown if the downstream measurement was not significantly lower than the upstream measurement at the 95% confidence level as determined by a one-tailed Student's t test. Calculated CADRs for each compound by experiment are presented in Figure 1. The effects of the parameters of concentration and air flow rate on the efficiency of the reactions for the individual compounds are generally apparent. Experiments 1 and 5 conducted at the low flow rate resulted in similar CADRs for most compounds, despite the two- to three-fold differences in inlet VOC concentrations between experiments.

Experiments 6 and 7 conducted at the high flow rate produced similar CADRs for a number of compounds, again with two- to three-fold differences in inlet VOC concentrations between experiments. Some compounds had notably higher average efficiencies in Experiment 7 with the lower inlet VOC concentrations. Particularly, the conversion efficiencies of ethanol, *tert*-butyl methyl ether (MTBE), isopropanol, 2-butanone, and carbon disulfide (CS2) were higher than expected. However, the uncertainties of the measurements of these very volatile compounds at low concentration were high, as shown by the error bars. Thus, a number of the differences were not significant. Overall, these results show that the fraction of an inlet VOC that reacts (i.e., pollutant removal efficiency) decreases approximately in direct proportion to increasing air flow rate and is less affected by the inlet VOC concentration.

Experiments with the synthetic office VOC mixture resulted in the net production of formaldehyde, acetaldehyde, formic acid, and acetic acid, which indicates incomplete decomposition of some of the VOCs in the inlet air stream. Additionally, there was no significant reduction in the acetone concentration in three experiments suggesting that acetone was a reaction product. As an example, Table 2 shows the inlet and outlet concentrations of formaldehyde, acetaldehyde, and formic and acetic acids from Experiment 6. The outlet concentrations of formaldehyde and acetaldehyde are 3.4 times and 4.6 times the inlet concentrations, respectively. An approximate linear relationship was observed between the formaldehyde net production rate in µmoles per hour and the supply rate of total alcohols and glycol ethers in µmoles per hour [1].

Detailed results of experiments conducted with cleaning product and building product mixtures of VOCs are presented elsewhere [1]. For the cleaning product VOCs, which included 2-BE, d-limonene, other terpenes and terpene alcohols, reaction efficiencies ranged from 0.2 to 0.8 and tended to be higher with a lower air flow rate. For the building product mixture, reaction efficiencies ranged from non-significant in a few instances to 0.8, with most values above 0.3. The reaction efficiencies measured in experiments with the aldehyde mixture, ranged from 0.18 to 0.49 as shown in Table 3.

The pressure drop imposed by the UVPCO system at the highest air flow rates was only 33 to 35 Pa. For reference, total pressure drops in supply airstreams of HVAC systems are often larger than 750 Pa.

4. Discussion

The current study has addressed several key recommendations made in a recent review [6]. Specifically, it was recommended that research be conducted to investigate: a) performance at low-level concentrations of VOCs that are representative of indoor environments; b) the use of UVPCO in airstreams with typical mixtures of pollutants; and c) the potential formation of reaction by-products. Our selection of the study compounds was based upon knowledge of VOCs frequently encountered in indoor air. The mixtures were comprised so that the individual VOCs were present at realistic relative abundances. Also, the concentrations used in the experiments were typical of those in office buildings and residences.

The results obtained at low ppb concentrations generally support the theory regarding the relative reaction rates of different chemical classes of compounds. The oxidation rates of the represented chemical classes followed the approximate order: alcohols and glycol ethers > aldehydes, ketones, and terpene hydrocarbons > aromatic and alkane hydrocarbons > halogenated aliphatic hydrocarbons. A relationship between molecular weight and oxidation rate was less apparent in our data.

The relative conversion rates of the various chemical classes within the UVPCO generally are favorable with respect to the use of the device for the treatment of indoor air in office buildings, schools, and residences. In a review of VOC concentrations measured in North American buildings since 1990, [2] it was shown that the concentrations of chlorinated solvents such as 1,1,1-trichloroethane, trichloro-ethene, and tetrachloroethene in residences have decreased relative to measurements made in the preceding decade. This change likely was due to the phasing out of the use of these chemicals in products in response to the U.S. 1990 Clean Air Act Amendments. As a result, VOC measurements made in recent years in houses, schools, and office buildings rarely show these chemicals above low background levels [7,8,9]. Indoor air concentrations of aromatic hydrocarbon solvents likely are decreasing for the same reason. Contemporary products often use alcohols, glycol ethers, and terpene hydrocarbons as substitute solvents [10]. Some of these oxygenated solvents that can occur at relatively high indoor concentrations have relatively low odor thresholds or chronic toxicity exposure guideline concentrations [11]. As a result, the high conversion efficiencies observed for alcohols, glycol ethers, and other oxygenated chemicals are well matched to the composition of chemicals of concern in indoor air. Conversely, the relatively poor performance observed for halogenated hydrocarbons is not viewed as a serious detriment.

If UVPCO is sufficiently effective, it may enable reductions in the rates of outdoor air supply. In a typical office installation of a UVPCO system in the supply air stream, overall VOC conversion efficiencies greater than approximately 17% are required to counteract the predicted concentration increases from a 50% reduction in ventilation. For the prototype UVPCO device evaluated here, the conversion efficiencies of nearly all VOCs exceed 17%, and the conversion efficiencies for many compounds of potential concern are well above 17%.

This study has generated substantial data on the production of gas-phase byproducts when a UVPCO is operated with indoor relevant mixtures of VOCs at realistic concentrations. Formaldehyde, acetaldehyde, acetone, formic acid, and acetic acid were produced in these experiments. No other significant byproducts were identified by the sampling and analytical methods employed in the study. Formaldehyde and acetaldehyde are recognized as important indoor air toxicants. They are categorized as carcinogens on the State of California Clean Water and Drinking Act of 1986 list of toxicants [12]. Recently, the International Agency for Research on Cancer has classified formaldehyde as a human carcinogen [13]. The California Office of Environmental Health Hazard Assessment (OEHHA) has developed acute and chronic exposure guidelines for formaldehyde exposure among the general population including sensitive individuals. The one-hour acute Reference Exposure Level (REL) is 74 ppb [14]. The long-term (i.e., 10 years or more) chronic REL is 2.4 ppb [14]. The California Air Resources Board's recommended guideline for formaldehyde concentrations in occupied buildings is 27 ppb, a value derived from the acute REL assuming an eight-hour exposure period [15]. The NIOSH guideline for formaldehyde in work environments including office buildings is 16 ppb [16]. The OEHHA chronic REL for acetaldehyde is 5 ppb [14]. These governmental agency guidelines suggest that indoor concentrations of formaldehyde and acetaldehyde should be maintained at very low levels. The other reaction products are of lesser concern. However, the 100% odor threshold detection level for acetic acid is 10 ppb [17].

The UVPCO produced formaldehyde and acetaldehyde as the result of incomplete decomposition of the reactants, but the UVPCO system also destroys a fraction of the inlet formaldehyde and acetaldehyde. A central question then is: "in an office building with air recirculated through a UVPCO system, what is net impact of UVPCO operation on indoor formaldehyde and acetaldehyde concentrations?" Based on preliminary modeling using the results of this study, we estimate roughly three-fold increases in indoor formaldehyde and acetaldehyde concentrations with UVPCO operation in an office building [1]; however, the actual increases will vary depending on the concentrations and composition of VOCs in the building.

5. Conclusions

The VOC conversion efficiencies achieved with the prototype device studied here suggest that a UVPCO air cleaner for gaseous contaminants may be beneficial for the large-scale treatment of air in occupied buildings and may ultimately allow for a reduction of outside air supply in offices and other buildings as an energy conservation measure. However, several issues remain to be investigated. In particular, the impact of UVPCO operation on formaldehyde and acetaldehyde concentrations inside buildings should be better quantified. Our initial calculations indicate a need to reduce the formaldehyde and acetaldehyde production rates in the UVPCO system or to combine UVPCO with some other air cleaning system capable of removing formaldehyde and acetaldehyde.

6. Acknowledgments

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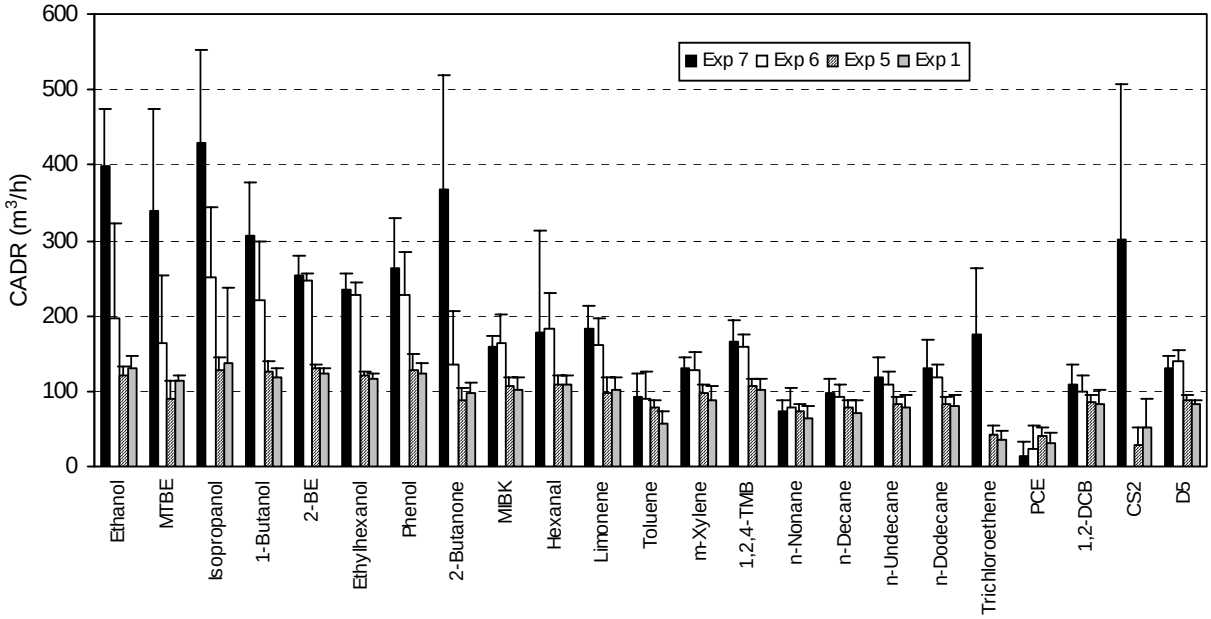

Figure 1. Clean air delivery rates (CADRs) of VOCs in four experiments with synthetic office VOC mixture (mean ± 1 stdev). Acetone, trichlorotrifluoroethane (R-11), dichloromethane (DCM), and 1,1,1-trichloroethane (1,1,1-TCA) showed insignificant upstream - downstream differences and were omitted. In Exps. 7 and 6, UVPCO was operated at ~580 m^3/h; in Exps. 5 and 1, UVPCO was operated at 165 – 171 m^3/h. Inlet VOC concentrations were lowest in Exp. 7, highest in Exp. 1., and intermediate in Exps. 6 and 5. MIBK=methyl isobutyl ketone; 1,2,4-TMB=1,2,4-trimethylbenzene; PCE= tetrachloroethene; 1,2-DCB=1,2-dichlorobenzene; D5=decamethylcyclopentasiloxane; other compounds defined in text.

Table 1. Inlet mixing ratios (ppb) and fractions of compounds reacted (mean ± 1 std dev) in four experiments with synthetic office VOC mixture. See text and Figure 1 for compound definitions.

Compound	***Exp 7 - Low Conc 581 m^3/h***		***Exp 6 - Mid Conc 579 m^3/h***		***Exp 5 - Mid Conc 171 m^3/h***		***Exp 1 - High Conc 165 m^3/h***	
	Mix Ratio (ppb)	*Fraction Reacted*	Mix Ratio (ppb)	*Fraction Reacted*	Mix Ratio (ppb)	*Fraction Reacted*	Mix Ratio (ppb)	*Fraction Reacted*
Ethanol	15.9±0.8	*0.69±0.07*	36±3	*0.34±0.11*	80±2	*0.71±0.03*	134±1	*0.80±0.05*
Isopropanol	10.5±0.8	*0.74±0.11*	27±1	*0.44±0.01*	41±2	*0.75±0.05*	81±19	*0.83±0.30*
1-Butanol	1.24±0.05	*0.53±0.06*	3.2±0.2	*0.38±0.07*	4.3±0.1	*0.73±0.04*	12.9±0.2	*0.72±0.03*
Ethylhexanol	1.84±0.03	*0.40±0.02*	4.5±0.1	*0.39±0.01*	5.4±0.1	*0.71±0.01*	12.0±0.1	*0.70±0.02*
Phenol	0.97±0.05	*0.45±0.06*	1.98±0.08	*0.39±0.05*	2.0±0.1	*0.75±0.06*	4.8±0.1	*0.75±0.04*
2-BE	2.8±0.1	*0.44±0.02*	6.9±0.1	*0.43±0.01*	7.1±0.06	*0.76±0.01*	19.4±0.1	*0.75±0.02*
MTBE	2.8±0.3	*0.58±0.12*	8.4±0.2	*0.28±0.08*	9.5±0.4	*052±0.07*	26	*0.68*
Acetone	14.0±0.5	*0.51±0.07*	32±2	*Ns**	45±4	*Ns*	103±6	*Ns*
2-Butanone	0.95±0.09	*0.63±0.13*	3.0±0.1	*0.23±0.06*	3.7±0.1	*0.52±0.05*	9.7±0.1	*0.59±0.05*
MIBK	2.7±0.1	*0.27±0.01*	7.0±0.2	*0.28±0.03*	8.8±0.1	*0.63±0.03*	23±1	*0.62±0.05*
Hexanal	0.96±0.07	*0.31±0.12*	2.1±0.1	*0.32±0.04*	2.3±0.1	*0.63±0.03*	7.5±0.1	*0.66±0.03*
Limonene	1.65±0.04	*0.32±0.03*	4.1±0.1	*0.28±0.03*	3.7±0.2	*0.57±0.06*	12.4±0.3	*0.62±0.05*
Toluene	9.2±0.2	*0.16±0.03*	22±1	*0.15±0.03*	26±1	*0.45±0.03*	54±1	*0.35±0.05*
m-Xylene	3.0±0.1	*0.23±0.01*	7.1±0.1	*0.22±0.02*	8.3±0.1	*0.57±0.03*	20±1	*0.54±0.06*
1,2,4-TMB	0.89±0.02	*0.29±0.02*	2.1±0.1	*0.28±0.01*	2.5±0.1	*0.62±0.03*	5.9±0.1	*0.62±0.04*
n-Nonane	1.92±0.01	*0.13±0.01*	4.9±0.1	*0.13±0.02*	6.0±0.1	*0.43±0.03*	15.5±0.2	*0.38±0.05*
n-Decane	1.77±0.03	*0.17±0.02*	4.4±0.1	*0.16±0.02*	5.4±0.1	*0.46±0.03*	12.8±0.1	*0.43±0.05*
n-Undecane	1.56±0.03	*0.20±0.02*	3.9±0.1	*0.19±0.01*	4.7±0.1	*0.49±0.02*	10.4±0.1	*0.48±0.05*
n-Dodecane	4.3±0.1	0.22±0.03	10.6±0.1	0.20±0.01	12.6±0.2	0.49±0.02	24±1	0.50±0.04
R-11	2.2±0.1	0.34±0.05	6.3±0.2	Ns	6.5±0.7	Ns	21	0.28
DCM	9.9±0.3	*0.23±0.09*	25±1	*Ns*	31±1	*Ns*	83±2	*Ns*
1,1,1-TCA	4.6±1.0	*0.51±0.24*	16.7±0.6	*Ns*	17.2±2.0	*Ns*	52	*0.45*
Trichloro-ethene	0.69±0.02	*0.30±0.07*	1.88±0.11	*Ns*	2.3±0.1	*0.25±0.03*	6.6±0.1	*0.21±0.04*
PCE	1.77±0.02	*≤0.05*	4.5±0.1	*≤0.05*	5.4±0.1	*0.23±0.03*	14.2±0.1	*0.19±0.04*
1,2-DCB	0.55±0.01	*0.19±0.02*	1.38±0.01	*0.17±0.02*	1.66±0.02	*0.50±0.03*	3.7±0.1	*0.51±0.03*
CS2	0.94±0.14	*0.52±0.18*	2.5±0.5	*Ns*	3.7±0.1	*0.16±0.07*	10.1±0.4	*0.32±0.12*
D5	0.64±0.01	0.22±0.02	1.62±0.01	0.24±0.01	1.95±0.01	0.52±0.02	4.7±0.1	0.50±0.02

Table 2. Data from Exp. 6. showing net production of formaldehyde, acetaldehyde and carboxylic acids.

Compound	**Mixing Ratio (ppb)**	**Mixing Ratio (ppb)**	**Ratio (inlet/outlet)**
Formaldehyde	3.0±0.2	10.0±0.8	3.4
Acetaldehyde	1.39±0.02	6.4±0.7	4.6
Formic acid	3.8±0.5	12.6±1.5	3.3
Acetic acid	7.8±0.7	17.6±2.1	2.2

Table 3. Inlet VOC mixing ratios (ppb) and fractions reacted (mean ± 1 std. deviation) in two experiments with mixture of formaldehyde and acetaldehyde.

Compound	***Exp 24 (280 m^3/h)***		***Exp 23 (167 m^3/h)***	
	Inlet Mix Ratio (ppb)	*Fraction Reacted*	Inlet Mix Ratio (ppb)	*Fraction Reacted*
Formaldehyde	25±3	*0.28±0.12*	33±2	*0.49±0.06*
Acetaldehyde	5.3±0.6	*0.18±0.11*	8.7±0.7	*0.44±0.09*