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Integrated Technology Air Cleaners (ITAC): Design and Evaluation

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Environmental Energy Technologies Division

September 2013

DISCLAIMER

Acknowledgements

PREFACE

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Integrated Technology Air Cleaners (ITAC): Design And Evaluation is the final report for task 2.1 of the Lawrence Berkeley National Laboratory Energy Efficiency Research Projects project (contract number 500-10-052) conducted by Lawrence Berkeley National Laboratory. The information from this project contributes to Energy Research and Development Division's Buildings End-Use Energy Efficiency Program.

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ABSTRACT

The primary objective of this project was to design, build, and test an air cleaner for residential use with the potential to substantially improve indoor air quality, or maintain indoor air quality unchanged, when outdoor air ventilation rates are reduced to save energy. Two air cleaners were designed and fabricated. The design targets for airflow rate, fan power, and projected cost were met. In short term laboratory studies, both units performed as expected; however, during field studies in homes, the formaldehyde removal performance of the air cleaners was much lower than expected. In subsequent laboratory studies, incomplete decomposition of some indoor air volatile organic compounds, with formaldehyde as a product of partial decomposition of volatile organic compounds, was confirmed as the explanation for the poor formaldehyde removal performance in the field studies. The amount of formaldehyde produced per unit of decomposition of other volatile organic compounds was substantially diminished by increasing the amount of catalyst on the filter and also by decreasing the air velocity. Together, these two measures reduced formaldehyde production, per unit destruction of other volatile organic compounds, by a factor of four, while increasing the removal efficiency of volatile organic compounds by a factor of 1.4. A company with a southern California office is conducting studies in conjunction with Lawrence Berkeley National Laboratory, with the goal of incorporating the ITAC catalytic air cleaning technology in their future commercial products.

Keywords: air cleaning, catalyst, energy efficiency, filtration, homes, ventilation

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EXECUTIVE SUMMARY

Introduction
Reductions in outdoor air ventilation rates of homes could save substantial energy. However, without compensating measures, indoor air quality will often be degraded posing health risks for the occupants of the home. Energy efficient air cleaning systems that are effective in removing particles and volatile organic compounds could enable reductions in ventilation rates and energy savings while maintaining or improving indoor air quality

Project Purpose
The primary objective of this project was to design, build, and test an air cleaner for residential use with the potential to substantially improve indoor air quality, or maintain indoor air quality unchanged, when outdoor air ventilation rates are reduced to save energy.

Project Results
Two air cleaners were designed and fabricated based on modifications of an existing commercially-available air cleaner. One unit, designated ITAC1, included a high efficiency particle filtration system and a catalyst-treated low-efficiency particle filter. The catalyst treated filter was included based on results of prior research showing that the catalyst was highly effective in destroying formaldehyde and moderately effective in destroying several other volatile organic compounds (VOCs). The second unit, designated ITAC2, added a filter containing granular activated carbon to the ITAC1 design. One additional air cleaner was designed, but not fabricated and tested because the design did not meet cost targets. This unit included a high efficiency particle filter system, a catalyst-treated low-efficiency particle filter, and an activated carbon fiber (ACF) cloth. The ACF cloth was effective in prior research in removing a broad spectrum of VOCs, but it needed to be regenerated each day using a small amount of heated air. The hardware for the regeneration process increased the projected cost of the air cleaner above the cost target.

The ITAC units provided or exceeded the target airflow of 71 L/s (150 cfm) at the medium and high fan speed settings with impressively low fan power (approximately 13 W at medium fan speed and 29 W at high fan speed). The goal was power consumption less than 60 W; thus, the ITAC units exceed this goal by a large margin. Both the ITAC1 and ITAC2 designs were based on easy modifications to an existing energy-efficient air cleaner with a known retail price. The incremental cost to modify the original products and mass produce air cleaners with the ITAC1 or ITAC2 design would be modest; thus, the anticipated final cost meets the cost target of $600 for a unit with a 71 L/s supply air flow rate.

In short term laboratory studies, both ITAC1 and ITAC 2 removed formaldehyde with approximately a 60% efficiency. ITAC1 removed several other VOCs with a 20% to 45% efficiency, while ITAC2 removed the same VOCs with a 40% to 90% efficiency.

The ITAC units were deployed in houses for approximately 40 day periods. They were operated most of the time, but turned off during some periods to obtain baseline data. Indoor and outdoor concentrations of VOCs, particles, and ozone were measured. A tracer gas system was employed to measure air exchange rates. During the field studies, the formaldehyde removal performance of the air cleaners was much lower than expected. Concentrations of particles decreased as expected. Indoor ozone concentrations were too low to enable an evaluation of the effect of air cleaner operation on ozone concentrations.

Incomplete decomposition of some indoor air VOCs, with formaldehyde as a product of partial VOC decomposition, was hypothesized as the explanation for the poor formaldehyde removal performance of ITAC1 and ITAC2 in the field studies. Laboratory studies confirmed that formaldehyde can be produced by incomplete decomposition of VOCs by the catalyst-treated filters. The amount of formaldehyde produced per unit of VOC decomposition was substantially diminished by increasing the amount of catalyst on the filter and also by decreasing the air velocity. Together, these two measures reduced formaldehyde production, per unit VOC destruction, by a factor of four, while increasing the VOC removal efficiency by a factor of 1.4. Limited testing indicated that substituting powdered activated carbon for a portion of the catalyst had a modest effect on VOC removal and formaldehyde production; however, this measure may be attractive because the powdered activated carbon will cost less than the catalyst. Results of mass balance modeling, described subsequently, suggest that a modified ITAC1 air cleaner, with a larger amount of catalyst or catalyst plus powdered activated carbon, and with a lower air velocity through the catalyst-treated filter, will simultaneously reduce indoor formaldehyde concentrations and concentrations of other VOCs when initial levels of VOCs other than formaldehyde are low or moderate. Model results suggested that increasing the amount of catalyst and decreasing the air velocity are not sufficient measures for homes with initially high concentrations of VOCs other than formaldehyde (i.e., strong indoor VOC sources), as formaldehyde concentrations may be increased during air cleaner use.

The plans for this research project included outreach to industry with the goal of identifying a company interested in further developing and marketing an air cleaner based on the ITAC design. The outreach effort was scaled back when it became evident from field study data that the ITAC units were not reducing formaldehyde levels as anticipated. However, several air cleaner manufacturers have expressed interest in incorporating filters, treated with LBNL's manganese oxide catalyst, in their products. One company, is actively evaluating catalyst-treated filters, and providing a modest amount of financial support to LBNL to facilitate the evaluations.

Project Benefits
The technical advances of this project, and the associated knowledge gained, bring us substantially closer to the goal of having a residential air cleaner that maintains or improves indoor air quality when outdoor air ventilation rates are reduced to save energy.

CHAPTER 1:
Project Objectives and Background

The primary objective of this project (Task 2.1 of "Lawrence Berkeley National Laboratory Energy Efficiency Research Projects", agreement 500-10-052) was to design, build, and test an air cleaner for residential applications with the potential to substantially improve indoor air quality, or maintain indoor air quality unchanged, when outdoor air ventilation rates are reduced to save energy. To meet this goal, the air cleaner must integrate technologies for removal of particles and gaseous pollutants into an effective, energy-efficient, reasonable-cost air cleaner. The technology addressed in this project was given the following name "Integrated-Technology Air Cleaner, abbreviated as "ITAC".

This project addresses two related problems. First, large reductions in the energy used for outdoor-air ventilation of buildings are necessary if California is to achieve its goals pertaining to reduced CO_2 emissions, zero-energy new buildings, and dramatically more energy efficient existing buildings. Second, exposures to indoor-generated air pollutants pose significant health risks. These problems are linked because reductions in outdoor-air ventilation rates to save energy will increase concentrations of indoor-generated air pollutants. While many types of pollutants are emitted from indoor sources, ventilation rates have a small impact on exposures to some types of pollutants, for example semi-volatile organic compounds (Parthasarathy, Chan et al. 2012). Also, many houses and most commercial buildings have insignificant indoor combustion-sources of inorganic gaseous pollutants, such as unvented combustion equipment and tobacco smoking. In California, radon sources are generally low, thus, the need to control indoor radon concentrations will rarely drive minimum ventilation requirements. Consequently, in a substantial fraction of California buildings, minimum ventilation requirements are driven by the need to control indoor concentrations of indoor-generated particles (some of which are bioaerosols), emitted from a variety of sources, and volatile organic compounds (VOCs), also from a variety of sources.

Particle filtration technologies are already mature and effective and can control indoor concentrations of indoor-generated particles at low cost (Fisk, Faulkner et al. 2002, Beko, Clausen et al. 2008). Particle filtration is already used routinely in commercial buildings to control indoor concentrations of particles and low efficiency filters are used in houses with forced air heating and cooling.

The options for controlling indoor concentrations of indoor-generated VOCs include ventilation with outdoor air, source control, and air cleaning. At present, ventilation is the primary measure employed to control indoor concentrations of VOCs; however, to enable energy savings other VOC control technologies are needed. While elimination of ventilation is not practical, reducing ventilation rates (e.g., by 50%) is a practical goal if other additional measures are employed to control indoor VOCs. Source control, which can be very effective and consume

no energy, is an excellent long term strategy; however, it is challenging to implement source control because of the large number of VOC sources, the limited information on VOC emission rates from sources, the frequent changes in building materials, equipment, and consumer products, and an incomplete understanding of which VOCs are important to health. This leaves air cleaning -- the focus of this project. In general, to enable reductions in ventilation rates and associated energy savings, VOC air cleaning methods must be effective for the spectrum of indoor-generated VOCs that have concentrations affected by ventilation rates and that are known or suspected to be important for health (Parthasarathy, Chan et al. 2012). While the identity of all important VOCs is uncertain, analyses are starting to identify the key VOCs, and these analyses indicate that the risks of formaldehyde will often be dominant among the chronic health risks of indoor VOCs (Logue, McKone et al. 2011, Logue, Price et al. 2012, Parthasarathy, Fisk et al. 2013). Analyses also indicate that particles in indoor air, arising from outdoor air and from indoor sources, likely pose the largest risks of chronic health effects from indoor air pollutants in homes (Logue, McKone et al. 2011, Logue, Price et al. 2012). Also, particles are carriers of allergens and inflammatory agents linked to allergy and asthma (IOM 1993, IOM 2000, IOM 2004).

If a building's ventilation rate is reduced, there is a decrease in the rate of removal of each indoor-generated air pollutant, causing indoor pollutant concentrations, for pollutants from indoor sources, to increase. To prevent the indoor pollutant concentrations from increasing, an air cleaner must fully make up for the loss in pollutant removal by ventilation. For most air cleaners, the product of the rate of airflow through the air cleaner and the pollutant removal efficiency of the air cleaner must equal the reduction in ventilation rate. Consider, for example, a 400 m^3 house, and a reduction in ventilation rate by 39 L/s from 78 L/s to 39 L/s (approximately from 0.7 h^{-1} to 0.35 h^{-1}). To prevent an increase in indoor pollutant concentration when using an air cleaner with a 55% pollutant removal efficiency, the flow rate of air through the air cleaner must be 71 L/s (71 equals the 39 L/s reduction in ventilation rate divided by the efficiency of 0.55). If the indoor air is well mixed, for example due to frequent use of forced air heating and cooling systems, a single air cleaner may be appropriate. In other cases, two air cleaners each with an air flow rate of 35.5 L/s may be preferable. In practice, air cleaners are often installed where people spend much of their time, such as in family rooms and bedrooms.

Existing VOC air cleaning technologies are generally considered too expensive for widespread application in buildings. The dominant technologies are gas adsorption on granular activated carbon and various chemisorption technologies. They have been available for decades, but are deployed in only a tiny fraction of buildings. Activated carbon (Fisk 2007) can remove a broad range of VOCs; however, it is more cost effective for industrial applications with high VOC concentrations than for general use in buildings where VOC concentrations are much lower. Also, activated carbon has a low capacity for formaldehyde. The adsorption process is reversible, thus, one VOC can drive another type of VOC off the activated carbon unless the carbon media is replaced well before it becomes saturated with VOCs. Also water vapor, when humidity is high, and increases and air temperature, can cause VOCs to be desorbed from the activated carbon and released into the indoor air. The dominant chemisorption technology is a

granular activated alumina treated with a potassium- or sodium-permanganate. The chemisorption media reacts with VOCs susceptible to oxidation, removing them permanently, while consuming the media. This technology is highly effective for formaldehyde. However, the costs are high. Costs can be estimated using manufacturer's data on formaldehyde removal capacity (25 g of media are required to remove 1 g of formaldehyde), media cost (~$20 per kg), an engineering estimate of the costs of energy required to move air through the media, and assumed inlet formaldehyde concentrations. For example, the annual cost of removing all the formaldehyde from 1 m^3/s of air with an inlet formaldehyde concentration of 24 $\mu g/m^3$ was estimated to be approximately $500. Because VOC contaminants other than formaldehyde simultaneously consume the chemisorbent, actual costs will be considerably higher. For commercial buildings (a residential estimate is not available), the U.S. annual average cost of obtaining the same amount of formaldehyde removal with ventilation is $390 (Sidheswaran, Destaillats, et al. 2012) and, in California, costs will be lower because of the milder California climate. Ventilation has the advantage of simultaneously removing other indoor generated contaminants. Thus, this chemisorption technology does not appear cost competitive relative to ventilation, except potentially in severe climates.

Based on the prior discussion, to enable reductions in ventilation rates and associated energy savings with air cleaning, the technology should be effective for particles and for a broad spectrum of VOCs and remove these air pollutants at a sufficient rate to counteract the reduction in ventilation rate. An air cleaning technology that is effective across the relevant range of physical and chemical properties can be assumed effective for other VOCs with similar chemical and physical properties but currently not recognized as important. Effectiveness in removing formaldehyde from indoor air is critical. In some buildings, risks from formaldehyde dominate (Logue, Price et al. 2012) among the risks of VOCs and air cleaning solely for formaldehyde and particles may be sufficient to enable a reduction in ventilation. The 9 $\mu g/m^3$ reference exposure level for formaldehyde, provided by California's Office of Environmental Health Hazard Assessment, is among the lowest reference exposure level for common indoor air VOCs. Practical VOC air cleaning methods must also consume less energy than ventilation and be cost competitive. An advantage of air cleaning, relative to ventilation and indoor source control, is that air cleaning can also reduce exposures to outdoor air pollutants such as VOCs, particles, and ozone.

CHAPTER 2:
Methods

2.1   Air Cleaner Design

Design targets were selected based on review of existing technologies, calculations of the air cleaning capacity needed in houses to maintain IAQ when ventilation rates are decreased by approximately 50%, and a review of the costs of existing air cleaners. The initial design targets for ITAC (within the research proposal) included an air flow rate of 47 L/s (100 cfm), a removal

efficiency of 70% for a range of VOCs and for 0.3 micrometer size particles, a time-average power consumption less than 50 W, and an estimated product cost less than $400 with mass production. The targeted 47 L/s (100 cfm) airflow and pollutant removal efficiencies were indicated via mass balance modeling to be sufficient to substantially reduce indoor pollutant levels in a 370 m³ (13000 ft³) new house with a typical air exchange rate for a new house of 0.3 h⁻¹, or to enable air quality to be maintained or improved in such a house with substantial, e.g., 50%, reductions in outdoor air ventilation rates. Subsequently, the targeted capacity of the air cleaner was increased to assure the product would be effective in larger houses, in existing houses with higher ventilation rates, and in the event of lower pollutant removal efficiencies. The design target was changed to 71 L/s (150 cfm) airflow capability and 60W power consumption. Depending on the size of the home and its existing ventilation rate, smaller or larger air cleaners, or installation of multiple air cleaners would be appropriate. Based on a review of the prices of air cleaners on the market, the cost target was changed to $600. Existing quality residential air cleaners with 71 L/s (150 cfm) flow rates, good particle removal capabilities and little or no effectiveness for VOC removal currently sell for $600 or more.

2.1.1 Air Cleaning Technologies

The initial intent was to design an air cleaner that integrated an existing high efficiency particle filter with two VOC air cleaning technologies identified in prior research as promising. The promising VOC air cleaning technologies and example results of the prior research are described in the following two paragraphs and depicted in Figures 1 and 2.

- A new manganese oxide catalyst (Sidheswaran, Destaillats et al. 2011, Sidheswaran, Destaillats et al. 2011a) developed by Lawrence Berkeley National Laboratory (LBNL) was deposited on a fibrous particle filter, with a few grams of catalyst per square meter of filter media. The catalyst is a powder that adheres to the filter through Van der Waals forces and tests indicate that it stays attached to the filter. As air passed through the filter, a range of VOCs were removed catalytically. As shown in Figure 1, tests in the laboratory found that the system removed formaldehyde with a time-average efficiency of approximately 65% over a 2400 h (100 day) period, starting at approximately 80% efficiency and equal to about 50% efficiency after 2400 h. The catalyst was also effective for many other VOCs. There was a slow drop in VOC removal efficiency over time but the catalyst lifetime exceeded the expected replacement interval of 1000 to 2000 h for the catalyst-treated filter.

- In laboratory studies, an activated carbon fiber (ACF) cloth deployed in a VOC-laden airstream adsorbed a broad range of VOCs (Sidheswaran, Destaillats et al. 2012). Once every 12 hours, the adsorbed VOCs were driven off the ACF cloth using heated air vented to outdoors, making the ACF cloth ready to again remove VOCs from indoor air. The flow rate of heated regeneration air was approximately 1% of the flow rate of air being cleaned. The regeneration period was 15 minutes each 12 hours and the regeneration air temperature was 150 °C. Figure 2 provides an example of the VOC

removal performance of the ACF system over six cycles of air cleaning and regeneration. In this example, VOC removal efficiencies were above 80% for all VOCs except formaldehyde.

Figure 1: Long term removal of formaldehyde, acetaldehyde, acetone and other VOCs (O-VOC) with the manganese oxide catalyst applied to a particle filter (Sidheswaran, Destaillats et al. 2011, Sidheswaran, Destaillats et al. 2011a).
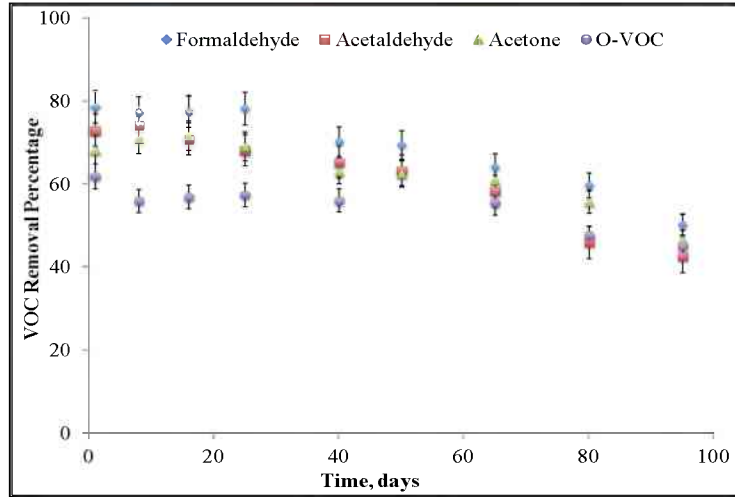
Figure 2: Example of VOC removal efficiencies of the ACF system (Sidheswaran, Destaillats et al.2012).
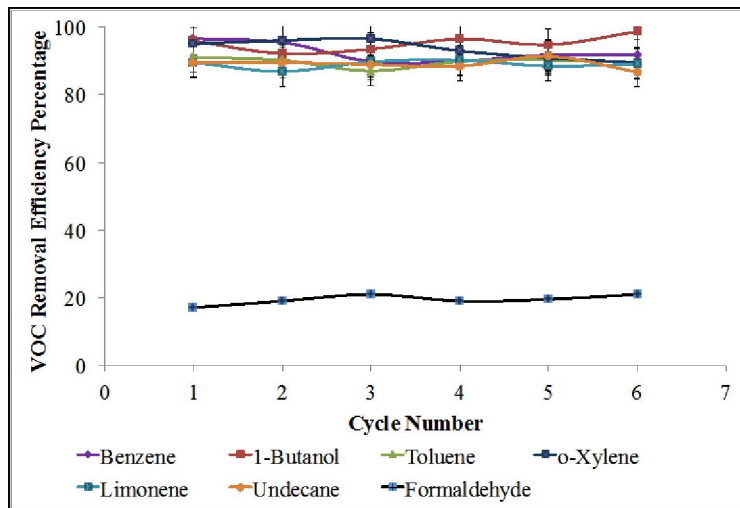
2.2   Characterization of Prototype Air Cleaners in the Laboratory

For each fan speed setting, air flow rates through the ITAC units were measured by injecting carbon dioxide into the air cleaner's inlet, measuring the carbon dioxide injection rates and the increases in outlet carbon dioxide concentration, and applying mass balance calculations. This procedure allowed measurement of flow rates without adding flow meters with airflow resistance that would change the rate of flow. Airstream pressure drops of the filter systems were measured with a sensitive pressure transducer and fan powers were measured with a sensitive watt meter.

Short-term studies, 24 to 120 h in duration, of the volatile organic compound (VOC) removal capabilities of the prototype air cleaners were completed with the units installed in a 27 m^3 stainless steel chamber. Air supplied to the chamber was filtered using a deep bed of activated carbon and a chemisorbent media to remove VOCs. Solutions of VOCs were evaporated into the indoor air at constant rates using syringe pump systems and heaters. Via these methods, formaldehyde and several other VOCs typical of indoor air VOCs, with a wide range of chemical properties, were added to the indoor air. To enable high measurement accuracy, initial concentrations of most individual VOCs were moderately elevated, relative to typical indoor air concentrations. After concentrations stabilized, ITAC 1 was turned on and inlet and outlet VOC concentrations were periodically measured over 120 h. Similar tests were performed with ITAC 2 over a 24 h period. Tests were performed using the high fan speed setting of the ITAC units, resulting in flow rates of approximately 78 L/s (165 cfm).

Integrated volatile carbonyl samples were collected upstream and downstream of the ITACs using dinitrophenyl hydrazine (DNPH)-coated silica samplers (Waters) at a rate of approximately 1200 cm^3 min^{-1} for 60 to 120 minutes. The flow corresponding to each sample was measured using a primary air flow calibrator (Gilibrator ®) with a precision greater than 2%. DNPH cartridges were extracted with 2-mL aliquots of acetonitrile, and the extracts were analyzed by high performance liquid chromatography with ultraviolet detection at λmax = 360 nm (Agilent 1200). A calibration curve for quantification was carried out using authentic standards of the formaldehyde-DNPH hydrazone. Quantification of volatile carbonyls was carried out following the United States Environmental Protection Agency Method T0-11A (http://www.epa.gov/ttnamti1/files/ambient/airtox/to-11ar.pdf)

Other VOCs were collected in multi-bed sorbent tubes with 70% bed of Tenax TA® sorbent backed with a section of Carbosieve ® (30%). Prior to use, the sorbent tubes were conditioned by helium purge (~10 cm^3 min^{-1}) at 310 °C for 60 minutes and sealed in Teflon capped glass tubes. Sampling flow rates were monitored periodically and samples were collected at approximately 100 cm^3 min^{-1} for a period of 1 hour so that the net sample volume was approximately 5L. VOCs were qualitatively and quantitatively analyzed by thermal desorption-gas chromatography/mass spectrometry generally following U.S. EPA Method TO-17 (http://www.epa.gov/ttnamti1/files/ambient/airtox/to-17r.pdf). For quantitative and qualitative

analysis of the target compounds, multi-point internal standard calibrations were created using pure compounds and 1-bromo-3-fluorobenzene as the reference compound.

Duplicate samples for volatile carbonyls and for others VOCs were collected upstream and downstream of the ITAC units. The differences among the replicate measurements were used to indicate measurement precision, communicated using error bars on figures. When there was a sampling or analysis failure in one on the duplicate samples, error bars are not displayed.

Single pass VOC removal efficiencies (ε) were calculated from the data using equation 1

$$\varepsilon = 100\% \, \frac{c_{in} - c_{out}}{c_{in}} \qquad\qquad 1$$

where C_{in} is the concentration of VOC in air entering the air cleaner and C_{out} is the concentration of VOC in air exiting the air cleaner. Propagation of error methods were employed to estimate uncertainties in removal efficiencies from the uncertainties in the measured VOC concentrations.

2.3   Tests of Prototype Air Cleaners in Houses

Two prototype air cleaners were installed in houses, prescreened to assure that indoor VOC concentrations were sufficiently high for accurate measurements. House 1 (H1) was a two story single family house with a floor area of approximately 180 m². House 2 (H2) was a single story apartment with one bedroom plus a den, and a floor area of approximately 80 m². Painting and cleaning in an adjacent apartment was an episodic source of VOCs in H2. In typical practice, it may be more common to install only one air cleaner in a home the size of H2.

Over approximately 40-day periods of air cleaner deployment, VOC concentrations were measured periodically at the inlet and outlet of air cleaners during a sample of days with the air cleaner operating and during a sample of days with the air cleaner not operating. When the air cleaners were operating, the high fan speed setting was used, resulting in flow rates of approximately 78 L/s (165 cfm). Two air cleaners were operated simultaneously in H1. In H2, one of two air cleaners was not operating during a portion of the field study.

The VOC sampling and analysis methods were as described above for the laboratory studies. In H1, airborne particle counts were measured indoors in the same room as the air cleaner, and outdoors, using Met-One Model BT-637 optical particle counters. These instruments count particles with diameters larger than 0.3 μm. In H2, particle concentrations were measured indoors in the same room as the air cleaner and outdoors using TSI Dustrak 2 instruments that produce an estimate of particle mass concentrations for particles less than 2.5 μm in diameter based on a light scattering technique. Particle measurements were not performed for the full 40 day periods due to competing demands for the instruments. Concentrations of particles were not measured at the outlet of air cleaners because of the anticipated measurement errors

associated with particle sampling from the high-velocity airstreams. To provide an indication of the effects of natural variations in house ventilation rates on VOC concentrations, a perfluorocarbon tracer gas (perfluoro-1,2-dimethylcyclobutane) was emitted continuously in the houses. Four vials were deployed at different locations in each house. To minimize any temporal variability in tracer gas emission rates, vials were installed in devices that maintained their temperature constant. Tracer gas diffused from the vials into indoor air through silicone septa, and tracer gas emission rates were based on the change in weights of the vials. Time average air exchange rates were estimated from a stead state mass balance calculation using the tracer emission rates and the measured tracer gas concentrations. Samples of air containing tracer gas were collected in multilayer gas sampling bags (Calibrated Instruments, Inc.) coincident with the periods of VOC sampling. The concentrations of tracer gas in these samples were analyzed using a gas chromatograph with electron capture detector calibrated with primary standard calibration gases. At each house, ozone concentrations were measured outdoors and at one indoor location using factory-calibrated 2B Tech Model 205 ozone monitors. The ozone measurements were made over approximately one-hour periods at times of VOC sample collection.

2.4   Evaluation of strategies for increasing contaminant decomposition

As discussed subsequently, data from the tests of air cleaners deployed in H1 and H2 suggested that some VOCs were not fully decomposed by the catalyst and that formaldehyde was a product of incomplete decomposition. Thus, the performance of the air cleaners was considered unsatisfactory and a set of subsequent laboratory studies (not include in the original scope of work) was undertaken to test this hypothesis and to evaluate technical options for reducing formaldehyde generation as a result of incomplete decomposition of VOCs. The intent of each option was to increase the time of contact of VOCs with the catalyst in order to more fully decompose the VOCs. The options were:
1. increase the amount of catalyst on the filter substrates;
2. decrease the air velocity; and
3. deploy a mixture of powdered activated carbon, which adsorbs many VOCs, and catalyst on the filter substrates.

The powdered activated carbon mixed with the manganese oxide catalyst for some experiments was a coconut-shell based nano-particle "super activated carbon" (U.S. Research Nanomaterials, Inc.).

The tests used sections, called coupons, from a 2.5 cm thick filter media intended for use in roll filters. The roll filter media (American Air Filter, Roll-O-Matt Green) has an arrestance efficiency rating (ASHRAE 2012) of 70% to 75%. Catalyst, or catalyst plus activated carbon, was applied to the filters using a powder paint sprayer and sections of the filter media were cut out to create the coupons. The amount of catalysts or catalyst plus carbon applied was determined

from the weight before and after application of catalyst. Coupons with 10 cm² of exposed face area were installed in a filter holder with an inlet and outlet tube. In the experiments, air containing individual VOCs or mixtures of VOCs were drawn through the coupons for four days. Upstream and downstream samples were collected once per day and analyzed as described above to determine carbonyl and other VOC concentrations. Because the inlet air contained no formaldehyde, all formaldehyde in the downstream samples was attributable to incomplete decomposition of VOCs. The reported concentrations of VOCs and formaldehyde from the upstream and downstream samples are the averages from the four measurements, one from each test day. The experimental methods were very similar to those described in greater detail in (Sidheswaran, Destaillats et al. 2011).

To characterize rates of formaldehyde production from incomplete decomposition of other VOCs, formaldehyde yields were calculated from equation 2

$$Y_i = C_f^0 / (\Delta C_i) \tag{2}$$

where Y_i is the yield of formaldehyde from incomplete decomposition of VOC i or VOC mixture i, C_f^0 is the concentration of formaldehyde in the air exiting the coupon, and ΔC_i is the decrease in concentration of VOC i or VOC mixture i as air passes through the coupon.

In the laboratory studies, the air entering the coupons contained only about 1 ppb of formaldehyde and air passed once through the coupons. However, when air cleaners are deployed in buildings, the air entering the air cleaner will contain formaldehyde, this formaldehyde is partially removed by the catalyst, and indoor air is recirculated through the air cleaner. With data from the small scale laboratory studies as model inputs, a steady state mass balance model was used to provide estimates of the net effect of air cleaner operation on indoor VOC concentrations. The model, which assumes that formaldehyde is the only significant product of incomplete decomposition, is described in equations 3 through 5.

For VOC i or VOC mixture i, where i is not formaldehyde

$$C_i = \left(\frac{Q}{V} X_i + \frac{S_i}{V}\right) / \left(\frac{Q}{V} + \frac{F}{V} E_i\right) \tag{3}$$

where C_i is the indoor air concentration, Q is the outdoor air flow rate, V is the indoor volume, X_i is the outdoor air concentration, S_i is the indoor VOC emission rate, F is the rate of air flow through the air cleaner, and E_i is the VOC removal efficiency of the air cleaner.

If no air cleaner is present, and C_i, Q, and V are known, or typical values are assumed as model inputs, the indoor VOC emission rate is calculated from equation 4.

$$S_i / V = Q / V (C_i - X_i) \tag{4}$$

For formaldehyde

$$C_f = \left(\frac{Q}{V} X_f + \frac{S_f}{V} + \frac{F}{V} \sum_2^n Y_i E_i C_i\right)\Big/\left(\frac{Q}{V} + \frac{F}{V} E_f\right) \qquad (5)$$

where subscript f refers to formaldehyde. Implicit in equation 5 is an assumption that formaldehyde yields Y_i from incomplete decomposition of VOC i are not dependent on the simultaneous presence of other VOCs. Because this assumption may be invalid, most experiments were performed with mixtures of VOCs entering the coupons and the resulting formaldehyde yields are the net yields for the mixture. Another implicit assumption is that the formaldehyde yield is not dependent on C_i the inlet VOC or VOC mixture concentration. There is likely some dependence of yield on inlet concentrations; however, assuming that if the yield is not a strong function of the inlet concentration, equation 5 can be used to estimate indoor formaldehyde concentrations with an air cleaner operating.

CHAPTER 3:
Results

3.1   Primary Air Cleaner Designs

The activated carbon fiber filter was included in the original design concept for removal of VOCs other than formaldehyde. With the subsequent realization that the manganese oxide catalyst was effective in breaking down a range of VOCs in addition to formaldehyde, the activated carbon fiber subsystem was considered less critical. In addition, cost estimations indicated that meeting the cost target was unlikely if the air cleaner incorporated the activated carbon fiber system, largely because of the 150 °C regeneration process and high costs of actuated dampers with seals able to withstand these temperatures. Without the activated carbon fiber system, the air cleaner prototypes could be produced via straight forward modifications of an existing energy efficient air cleaner. The Rabbitair Minus A2 SPA-780A air cleaner, incorporating a stack of filters in series, was used as a starting point. This unit has a very energy efficient fan system with a brushless DC fan motor and it is one of the quietest air cleaners identified. In its original configuration, manufacturer's reported fan power is about 40W with a 71 L/s (150 cfm) airflow. The retail cost is $460.

Table 1 identifies the filter elements in the two primary air cleaner designs, denoted ITAC1 and ITAC2, and lists the types of air pollutants these filters are designed to remove. ITAC 2 differs from ITAC1 via substitution of filter element 2b for filter element 2a. Filter element 2a is the medium efficiency particle filter supplied with the RabbitAir unit. In contrast, filter element 2b (Figure 3) is a commercially-available filter pad (Trisorb III) containing 3.2 kg of activated carbon per square meter of filter (300 g per ft^2), sized to fit the filter housing. Activated carbon is effective in removing VOCs, particularly those with a high molecular weight; however,

calculations suggest that the amount of activated carbon in this filter is not sufficient for effective long-term VOC removal. Activated carbon has a very low capacity for removal of formaldehyde (Fisk 2007) but has been shown to be effective for an extended period in removing ozone. Filter element 2b was included in ITAC2 because it may keep some high molecular weight VOCs from reaching filter element 4. These high molecular weight VOCs might shorten the life of filter element 4 by deactivating the catalyst. Filter element 4 (Figure 4) is a low efficiency (MERV 7) pleated synthetic media filter, 2.54 cm deep (Tripleat ES40L), treated by LBNL with a new manganese oxide catalyst. Approximately 2g of catalyst has been deposited on each filter. The catalyst is a powder applied via a spray process. Figure 5 shows a photograph of ITAC1 and ITAC2, indistinguishable in external views. Table 2 provides the measured air flow rates, airstream pressure drops, and fan powers of the ITAC units. The air flow rates meet the design target. The fan power at a 78 to 79 L/s air flow rate, moderately above the target flow rate of 71 L/s , is only 29 W, which compares favorably to the targeted maximum fan power of 60W.

Table 1: Filter elements in ITAC1 and ITAC2.

Filter Element	Filter area (m^2) face area [total area]	Pollutants Removed	ITAC1	ITAC2
(1) washable coarse particle screen	0.14 [0.14]	large particles and fibers	✓	✓
(2a) medium efficiency particle filter	0.15 [0.15]	large and medium size particles	✓	
(2b) filter pad with embedded activated carbon	0.15 [0.15]	ozone, high molecular weight VOCs		✓
(3) high efficiency particulate air (HEPA)	0.14 [1.0]	particles of all sizes	✓	✓
(4) low efficiency particle filter treated with LBNL MnOx catalyst	0.13 [0.21]	aldehydes and other VOCs	✓	✓

Figure 3: Filter element 2b, a pad filter containing activated carbon.

Figure 4: Filter element 4, a low efficiency pleated particle filter after treatment with manganese oxide catalyst.

Figure 5: Photographs of the modified air cleaner. The sheet metal fitting at the top is a temporary feature installed to facilitate sampling from the outlet airstream.

Table 2: Air flow rates, pressure drops, and fan power consumption.

Unit	Fan Speed	Flow Rate L/s (cfm)	Pressure Drop Pa	Fan Power W
ITAC1	silent	22 (47)	19	6
	low	28 (59)	28	7
	medium	45 (96)	45	13
	high	78 (166)	73	29
	turbo	101 (215)	94	50
ITAC2	silent	23 (48)	20	6
	low	31 (66)	28	8
	medium	52 (110)	43	13
	high	79 (167)	68	29
	turbo	103 (219)	90	53

3.2   ITAC secondary design

Before selecting the primary ITAC designs for further evaluation, a design incorporating the activated carbon fiber technology was produced. This secondary design incorporates a particle filter, an activated carbon fiber filter and regeneration system, and a catalyst treated filter. Because of the high cost of actuated dampers that would maintain integrity at the 150 °C regeneration temperature, the secondary design incorporates a system for moving the activated carbon fiber filter into and out of an insulated compartment where regeneration takes place. This design is described in appendix A.

3.3   Tests of ITAC Prototypes in Laboratory

Based on tests in the laboratory, Figures 6 and 7 show the measured single-pass formaldehyde removal efficiencies of ITAC1 and ITAC2, respectively. The formaldehyde removal efficiency of ITAC 1 increased over the first 24 hours and stabilized at approximately 60%. The formaldehyde removal efficiency of ITAC2 stabilized more quickly at approximately 60%. In the prior small scale laboratory tests (Figure 1), initial formaldehyde removal efficiencies were almost 80%. The thicker filter media to which catalyst was applied in the prior laboratory studies may explain the higher initial formaldehyde removal efficiency.

Figure 6: Formaldehyde removal efficiency of ITAC1 in laboratory studies.
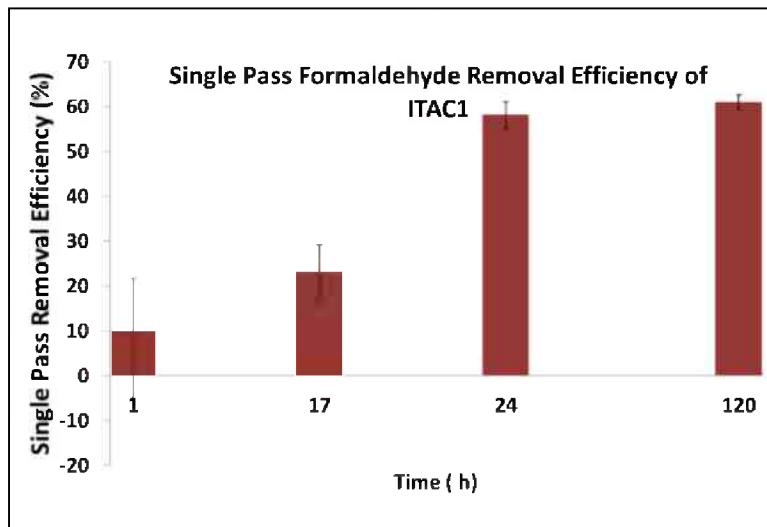
Figure 7: Formaldehyde removal efficiency of ITAC2 in laboratory studies.
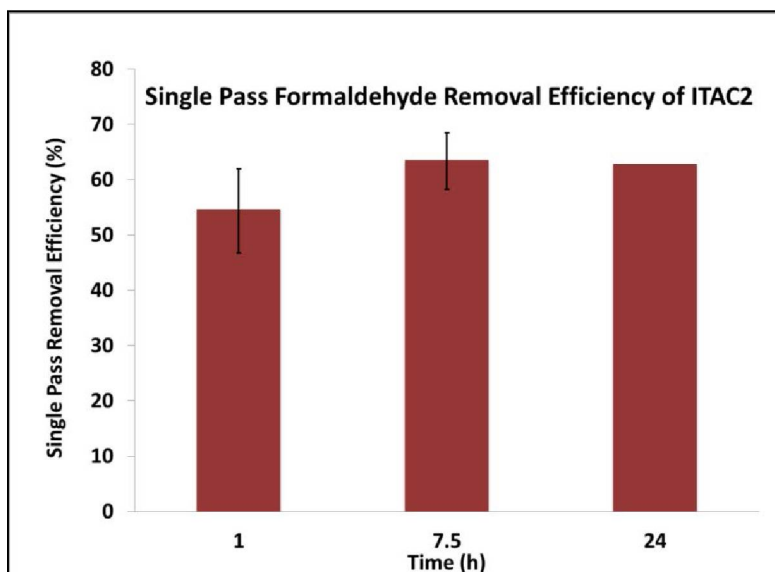
Figures 8 and 9 show single-pass removal efficiencies for other VOCs of ITAC1 and ITAC2, respectively. In some instances, the concentrations of individual VOCs were very low resulting in substantial uncertainties in single pass VOC removal efficiencies. In tests of ITAC 1, the VOC removal efficiencies of several of the VOCs were in the 30% to 40% range. Efficiencies for hexane, butanal, and benzene varied highly over time, and the variability is not fully attributable to the estimated measurement uncertainty. For most VOCs, efficiency appeared to increase over time. In tests of ITAC2, initial VOC removal efficiencies were approximately 40% for benzene and 80% to 90% for other VOCs. The higher initial VOC removal efficiencies of ITAC2 are likely a consequence of VOC adsorption on the activated carbon. With the limited quantity of activated carbon in ITAC2 it is likely that VOCs removal efficiencies for the lower molecular weight VOCs would not be maintained unless the carbon-containing filter was replaced frequently.

Figure 8: VOC removal efficiencies of ITAC1 in laboratory studies.
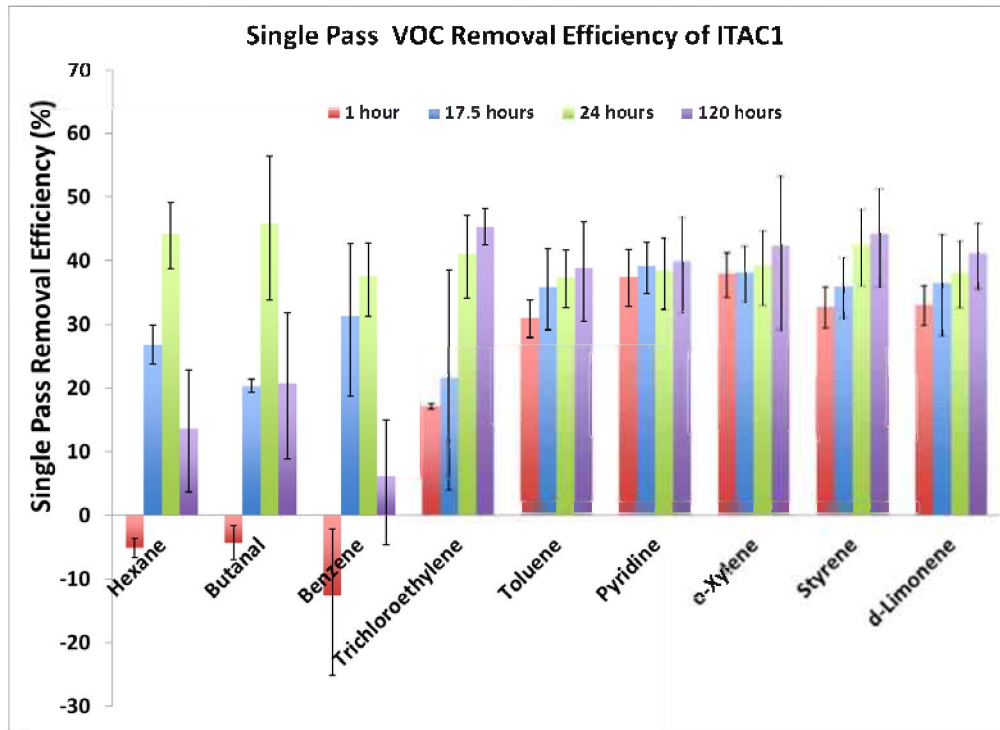
Figure 9: VOC removal efficiencies of ITAC2 in laboratory studies.
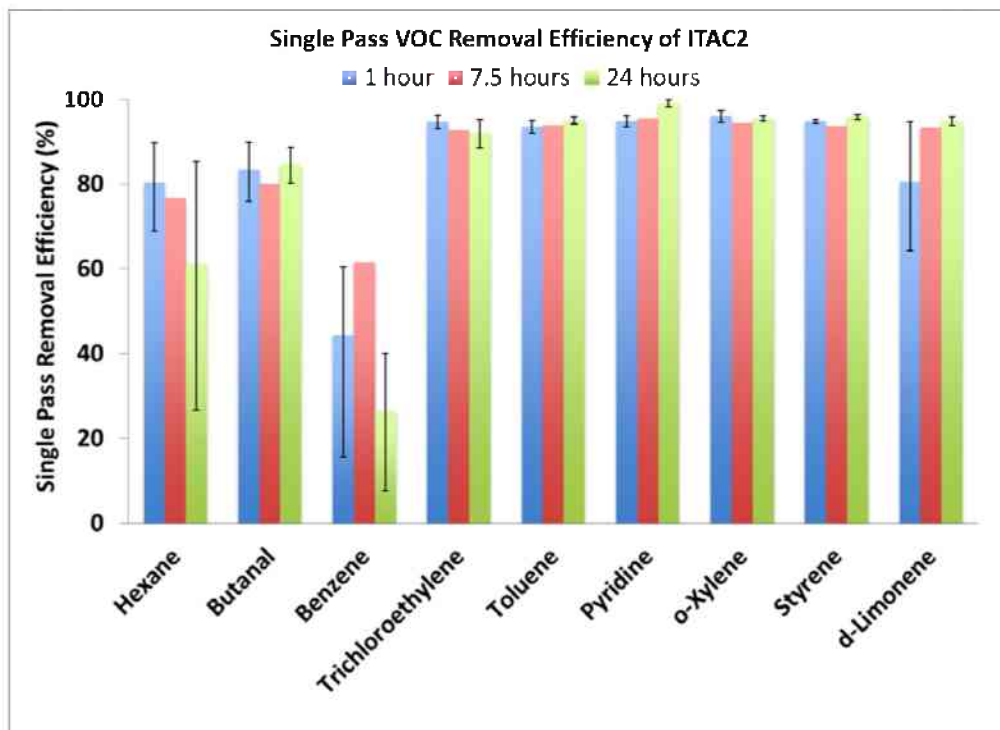
3.4   Tests of Prototype air cleaners deployed in houses

Figure 10 shows the measured VOC concentrations (excluding formaldehyde) in H1 with and without operation of two ITAC1 air cleaners, both installed in the same upstairs room. The door to this room was normally open. Indoor concentrations are smaller during periods of air cleaner operation, except concentrations on day 31, with the air cleaner operating are almost equal to concentrations on day 28 without air cleaner operation. Also shown is the tracer gas concentration. The moderate variability in tracer gas concentration over time is an indication of the moderate variability in house ventilation rates. Indoor air VOC concentrations on days 8 and 28, both with no air cleaner operation are very different, despite similar tracer gas concentrations indicating similar ventilation rates. These data indicate much higher indoor VOC emission rates on day 8, with an almost three-fold higher emission rate of styrene. Limonene concentrations, and thus emission rates are also several times higher on day 8 than on day 28. Given the variability in indoor VOC emission rates and, to a lesser extent, in ventilation rates, the evidence that the air cleaner lowered indoor VOC concentrations is not fully conclusive. On most days with the air cleaner operating, the concentrations of VOCs exiting the air cleaner are lower than the concentrations entering the air cleaner, indicating VOC removal by the air cleaner; however, the ratio of outlet to inlet concentration is highly variable and on two days exceeds unity.

The formaldehyde concentrations in H1 with and without operation of the ITAC1 air cleaners are shown in Figure 11. The average concentrations provided on this chart, and on Figure 12, are simple numerical averages of the concentrations measured at the inlet of the air cleaners. Indoor formaldehyde concentrations average 14 and 18 ppb in the two periods without ITAC operation and average 8 and 10 ppb in the two periods with ITAC operation. The decrease is significant but smaller than expected. During periods of ITAC operation, the formaldehyde concentration in the air exiting the air cleaner was typically about 15% to 20% lower than the inlet concentration, indicating a 15% to 20% single-pass formaldehyde removal efficiency, and on days 38 and 40 inlet and outlet concentrations are essentially identical indicating a 0% single-pass formaldehyde removal efficiency. Based on the prior laboratory studies (Figures 6 and 7) a much higher single-pass formaldehyde removal efficiency of 60% was anticipated. This discrepancy, and a similar one based on data from House 2 were suspected to be the consequence of incomplete decomposition of some of the VOCs passing through the air cleaner, with formaldehyde as a product of incomplete decomposition.

Figure 10: VOC data from field studies in House 1 with and without operation of ITAC1.
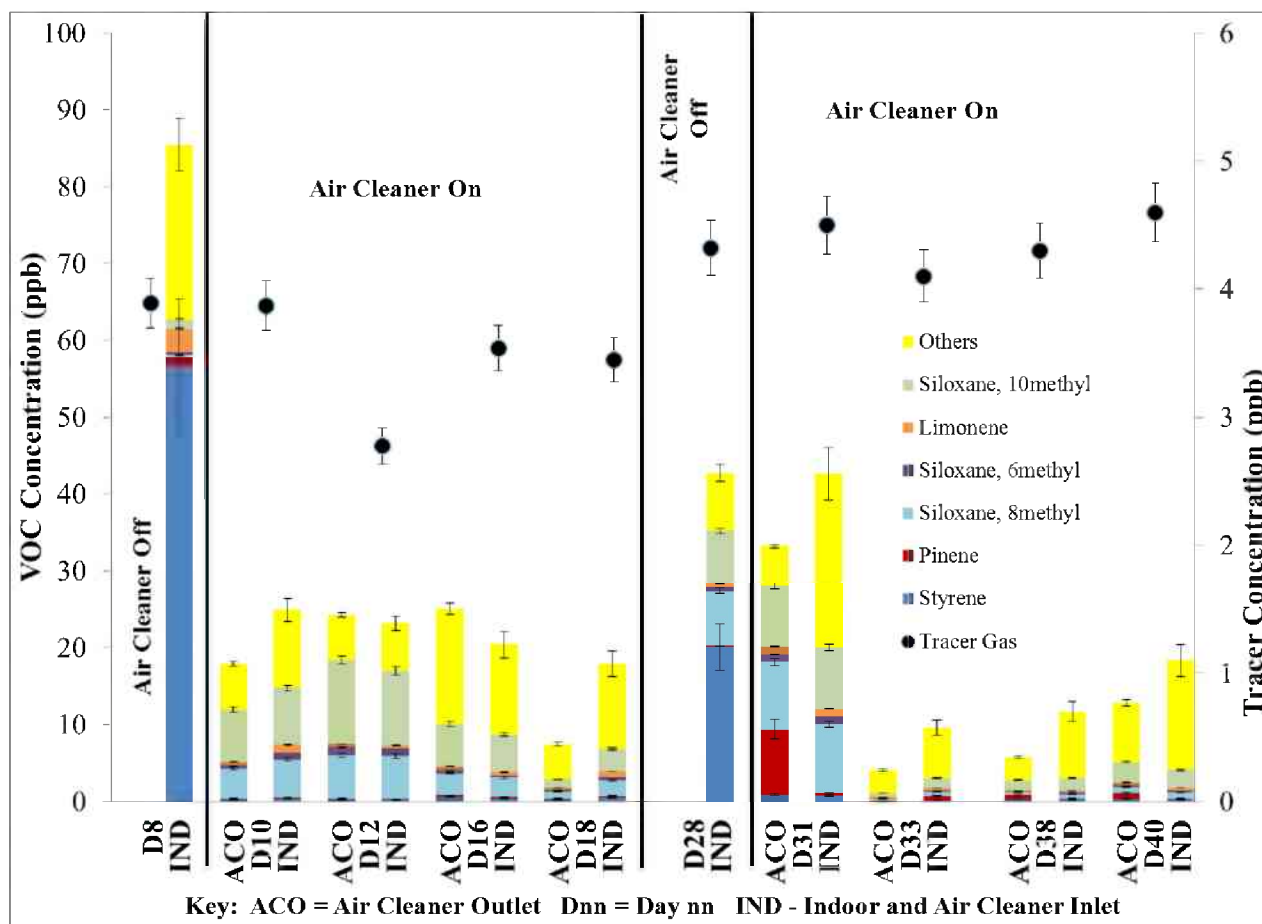
Figure 11: Formaldehyde data from field studies in House 1 with and without operation of two ITAC1 air cleaners.
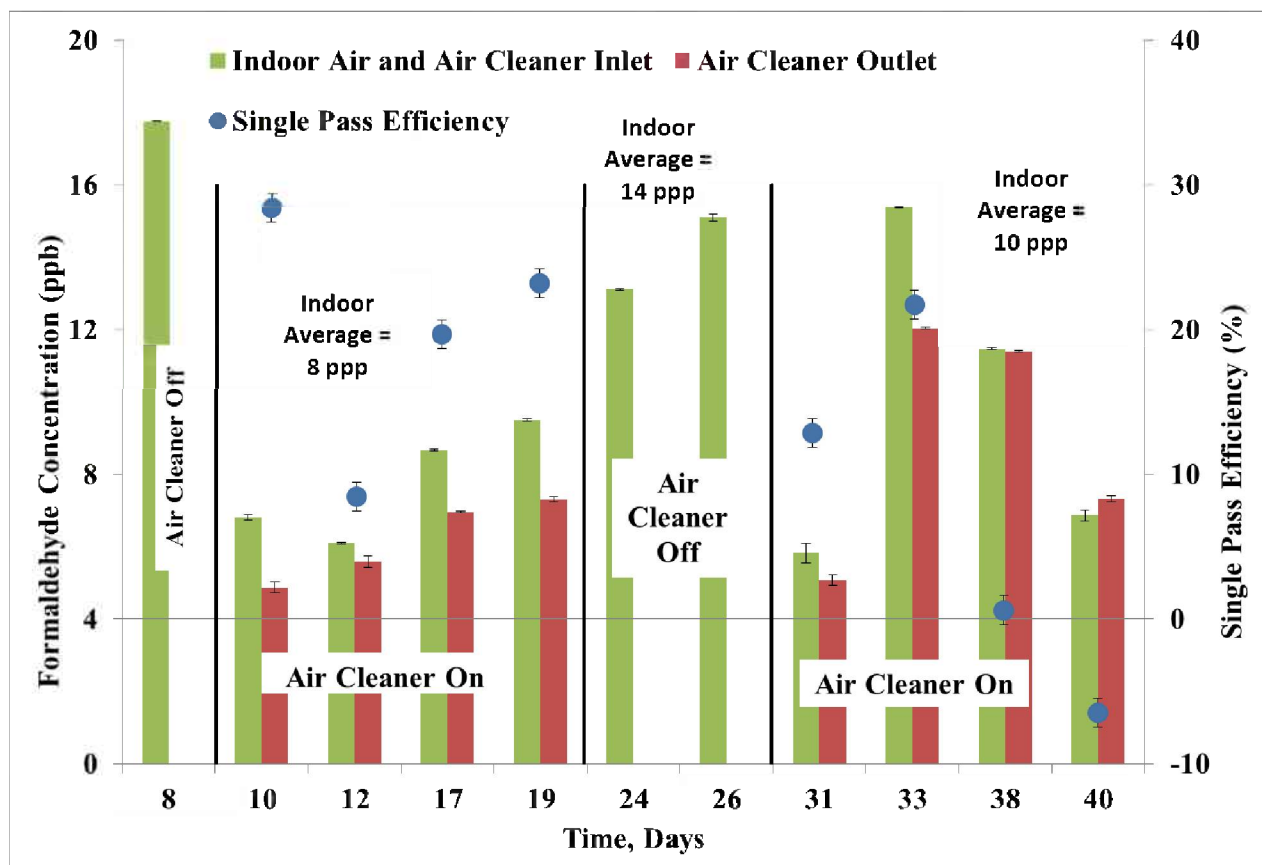
Figure 12 shows the results of VOC measurements (excluding formaldehyde) in H2 with and without ITAC2 operating. As in House 1, average indoor VOC concentrations are lower during the periods of ITAC operation. Substantial variations in VOC concentrations during periods of ITAC operation suggest considerable variability in indoor VOC emission rates, particularly for the "other VOC" category and for decamethyl-cyclopentasiloxane. Painting and cleaning in an adjacent apartment may have contributed to the temporal variability in VOC levels.

Concentrations of VOCs at the outlet of the air cleaner were typically significantly lower than inlet concentrations, indicating VOC removal by the air cleaner; however, the ratio of outlet to inlet concentrations was highly variable, indicating a highly variable VOC removal efficiency. On day 17, the total concentrations of VOCs exiting the air cleaner exceeded the inlet concentration. There was no clear evidence that ITAC2, with the activated carbon filter element was superior to ITAC1 without this element.

Figure 12: VOC concentrations from field studies in House 2 with and without operation of ITAC2.
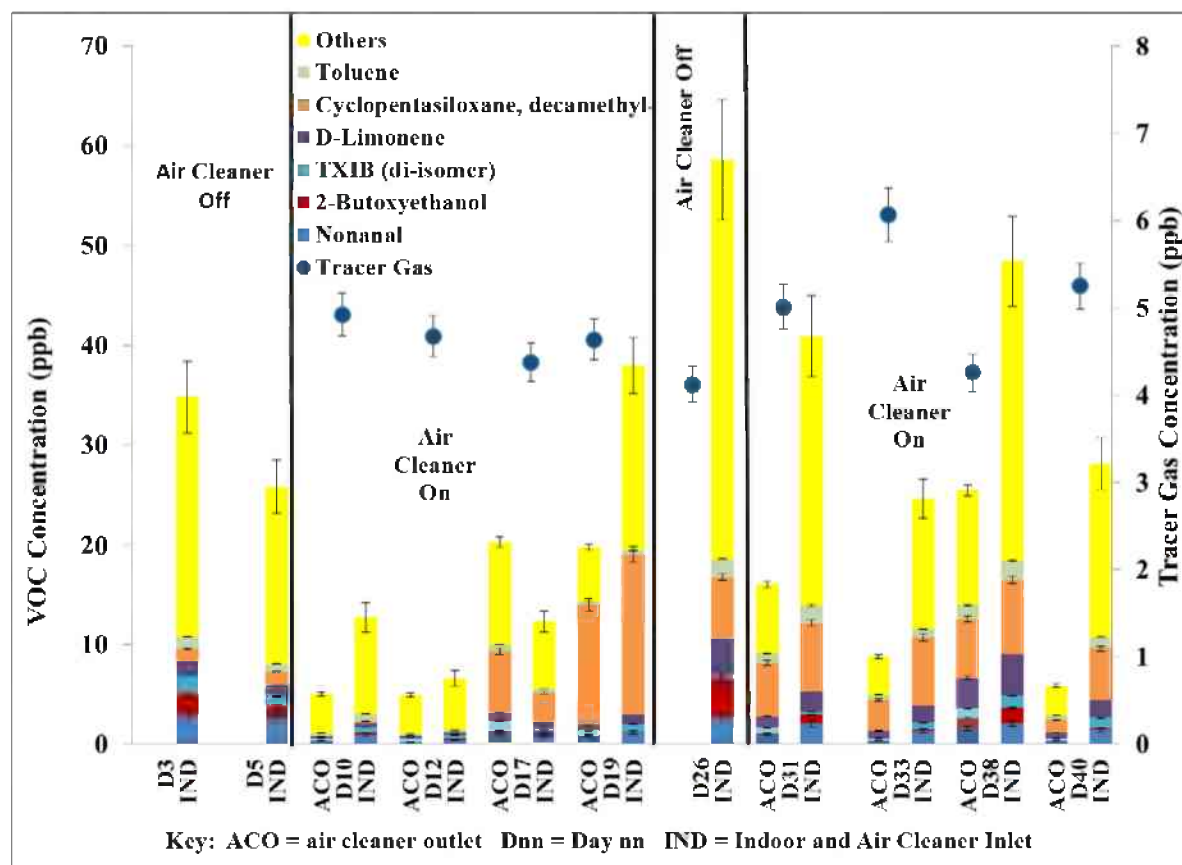
The results of the formaldehyde measurements in H2 are shown in Figure 13. Average indoor concentrations of 20 and 27 ppb during periods without ITAC operation are similar to average indoor concentrations of 14 and 31 ppb during periods of ITAC operation. Except on one day, formaldehyde removal efficiencies are 22% or lower and on day 40 the efficiency is essentially zero. Thus, the formaldehyde removal in the field studies is again far inferior to that measured in prior laboratory studies and these results suggest that formaldehyde is being produced from incomplete decomposition of some of the VOCs entering the air cleaner.

The tracer gas concentrations in Figures 10 and 12 indicate low air exchange rates. The air cleaner operation does not influence air exchange rate, thus the variability in tracer gas concentrations and associated ventilation rates must be a consequence of variability in weather conditions and occupants' use of doors and windows, although we requested that windows remain closed. For H1, average rates were 0.08 h^{-1} when the air cleaner operating, and 0.07 h^{-1} when the air cleaner was turned off. The corresponding air exchange rates for H2 were 0.13 h^{-1} and 0.15 h^{-1}, for periods with the air cleaner operating, and turned off, respectively, although only one measurement was available when the air cleaner was turned off. The low air exchange rates may be attributable to closed windows and mild summer weather.

Figure 13: Formaldehyde concentrations from the field studies in House 2 with and without operation of ITAC2.
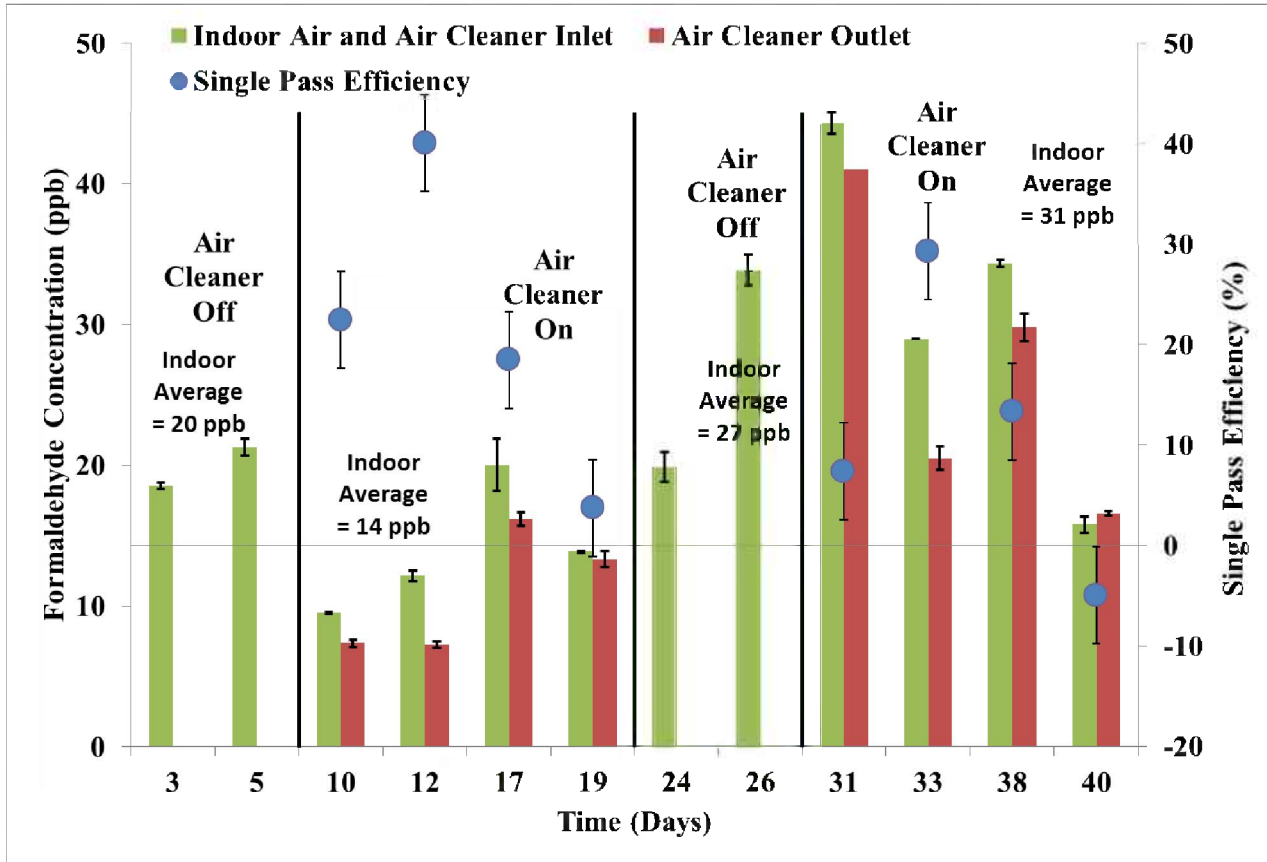
The results of the particle measurements in H1, with and without operation of two ITAC1 air cleaners, are shown in Figure 14. The indoor-to-outdoor ratios of particle counts are 0.2 or less during the 26 days of particle data collection with the air cleaner operating. In contrast, the corresponding ratios range from 0.9 to 1.3 during the seven days of data collection with the air cleaner turned off. The decrease in the indoor-to-outdoor particle concentration ratio is roughly 80% when ITAC1 units operate.

The results of the particle measurements in H2, with and without operation of ITAC2, are shown in Figure 15. In this house, there were periods with very high indoor particle concentrations, sometimes exceeding 1000 µg/m³ and correspondingly high ratios of indoor to outdoor particle mass concentrations. The tenant attributed the periods of high particle emission rates to cooking and cleaning. Excluding these time periods, the indoor-to-outdoor ratios of particle mass concentrations were typically 0.1 to 0.4 on days of air cleaner operation and 0.5 to 0.9 on days with the air cleaner turned off.

In H1 there is a very clear and large, approximately 80%, decrease in the ratio of indoor to outdoor particle count concentrations when the ITAC1 units operated. In House 2, the ratio of indoor to outdoor particle mass concentrations is lower during ITAC2 operation by roughly

50%, but only if one ignores the data from times when there is clearly a very strong indoor particle source. The two air cleaners should be about equally efficient in removing particles from the air passing through the air cleaners, as both included the same HEPA filter element and both had a very similar air flow rate. Given the smaller size of H2, a larger decrease in particle concentrations would be expected in H2 than in H1. The discrepancy between expectation and findings might be explained, in part, by imperfect mixing of indoor air. Also, particle count measurements (H1) are dominated by counts of small particles while particle mass measurements (H2) are strongly affected by large particles. Based on theory and prior empirical data (Fisk 2013), air cleaners with HEPA filters will be more effective in reducing indoor concentrations of small particles.

Figure 14: Indoor to outdoor ratios of particle count concentrations in House 1.
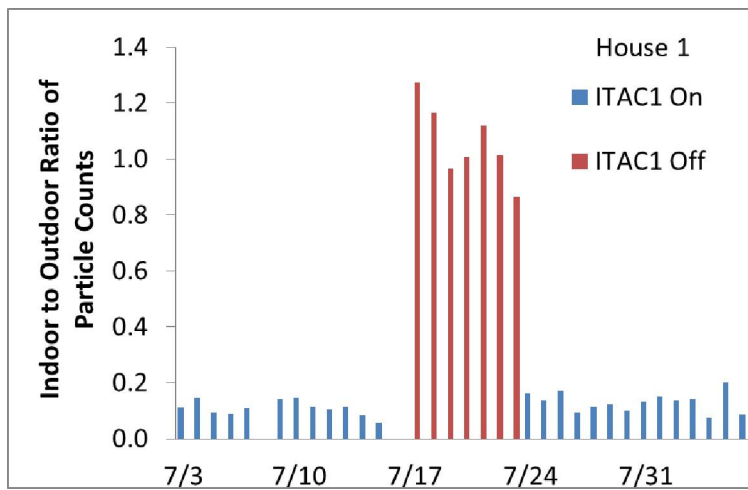
Figure 15: Indoor to outdoor ratios of particle mass concentrations, for particles smaller than 2.5 μm, in House 2.
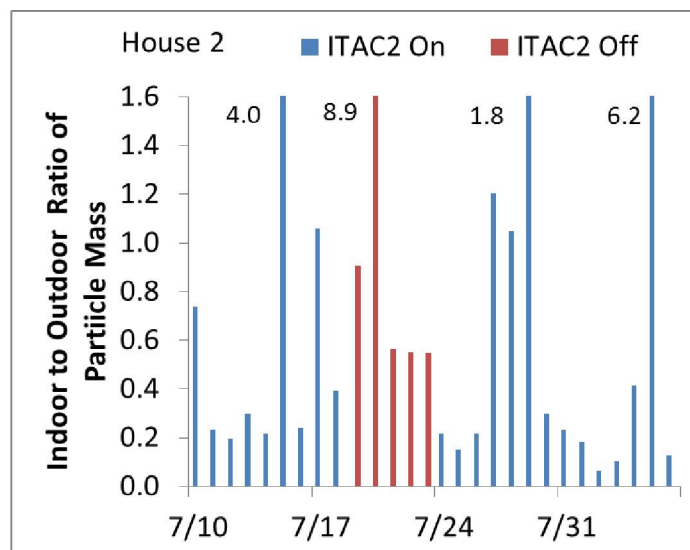
The large spikes in concentrations may be a consequence of cleaning and cooking events.

Indoor ozone concentrations in both houses were very low, normally below 1 ppb, with and without air cleaner operation. With such low concentrations, at the limit of measurement precision, the influence of air cleaner operation on indoor ozone concentrations could not be determined.

3.5 Evaluation of strategies for increasing contaminant decomposition

The results of experiments to quantify formaldehyde production rates from incomplete destruction of individual VOCs are shown in Table 3. With benzene, toluene, or o-xylene in the inlet air, 0.18 ppb of formaldehyde was produced per each 1 ppb of VOC destroyed. With styrene in the inlet airstream, 0.36 ppb of formaldehyde was produced per each 1 ppb of styrene destroyed. These data confirm that the system can produce formaldehyde from incomplete VOC decomposition. These findings support the hypothesis that incomplete VOC decomposition leading to production of formaldehyde explains why the formaldehyde removal efficiencies of the ITAC units during the field studies were less than expected.

Table 3: Results of tests with single VOCs in the inlet airstream, quantifying formaldehyde production rates.

Catalyst Loading (g/m^2)	Test VOC	Velocity (m/s)	Test VOC Inlet Concentration (ppb)	Test VOC Outlet Concentration (ppb)	Test VOC Removal Efficiency (%)	Outlet Formaldehyde Concentration (ppb)	Formaldehyde Yield [outlet formaldehyde ppb ÷ Δ test VOC ppb] (ppb/ppb)
30	benzene	0.5	24	16	32	1.4	0.18
30	toluene	0.5	31	20	35	1.9	0.18
30	o-xylene	0.5	38	24	35	1.9	0.18
30	styrene	0.5	37	26	29	3.8	0.36

In actual applications, the air entering an air cleaner will contain a mixture of VOCs and the amount of formaldehyde produced by the air cleaner will not necessarily equal the sum of the amounts of formaldehyde produced with each individual VOC alone entering the air cleaner. Consequently, Table 4 (columns 5 – 9) show the results of experiments performed with a mixture of VOCs, but no formaldehyde, entering the test system. In this table, the left set of nine columns contain experimental data with the data in columns 5-9 also serving as model inputs. The middle set of columns (10-15) are additional model inputs describing assumed house characteristics and the air cleaner's air flow rate. The right two columns are model predictions of percent reduction in indoor total volatile organic compound (TVOC) concentration and percent reduction in indoor formaldehyde concentration with air cleaning. The symbols for variables in the headings of columns, 5, 6, 10, 12-15, provide links to equations 2-5. Note that

there were 11 experiments, with 11 corresponding rows in the table, and that the 11 rows of experimental data are shown three times, for use in conjunction with different assumptions in columns 10-15 about the characteristics of the house and air cleaner air flow rates.

Consider first the experimental data, with the formaldehyde yield (ratio of formaldehyde produced to TVOC reacted and removed) as the key experimental result. Increasing the catalyst loading from 8 to 30 g/m^2 has the desired effect, decreasing the formaldehyde yield by a factor of 2 to 2.6, while also improving the TVOC removal efficiency when the air velocity is 0.17 m/s. Decreasing the air velocity from 0.5 to 0.17 m/s reduces the formaldehyde yield only slightly with 8 g/m^2 of catalyst on the filter, but reduces the yield by a factor of two with 30 g/m^2 of catalyst on the filter. Simultaneously increasing the catalyst loading from 8 to 30 g/m^2 and decreasing the air velocity from 0.5 to 0.17 m/s reduces the formaldehyde yield by a factor of four while increasing the TVOC removal efficiency by a factor of 1.4. However, increasing the inlet TVOC concentration from 40 to 103 ppb approximately doubled the formaldehyde yield based on a single pair of tests. Substituting powdered activated carbon for a portion of the catalyst did not dramatically change the amount of TVOC removed or the formaldehyde yield as long as the catalyst loading was at least 10 g/m^2. Because the activated carbon will be less expensive than the catalyst, applying a mixture of catalyst and powdered activated carbon to a filter may be more cost effective than just applying catalyst, with the mixture containing at least 10 g/m^2 of catalyst. One test with 15 g/m^2 of catalyst plus 15 g/m^2 of activated carbon, resulted in a low amount of TVOC removal out of alignment with the results of other tests with ten or more g/m^2 of catalyst

Table 4: Results of tests and model predictions with mixtures of VOCs in the inlet airstream, while varying catalyst loading, air velocity, and substituting activated carbon for a portion of the catalyst.

Experimental Data and Model Inputs									Other Model Inputs						Model Outputs	
Catalyst Loading (g/m²)	Carbon Loading g/m²	Carbon ÷ Catalyst	Velocity (m/s)	Inlet TVOC C_i (ppb)	TVOC Removal Efficiency [TVOC Reacted ÷ Inlet TVOC] E_i	Outlet Formaldehyde (ppb)	Formaldehyde Yield [Formaldehyde Produced ÷ ΔTVOC] (ppb/ppb)	Formaldehyde Produced ÷ Inlet TVOC (ppb/ppb)	Air Exchange Rate Q/V (h⁻¹)	Indoor Formaldehyde With No Air Cleaning (ppb)	Air Cleaner Flow Rate F/V (h⁻¹)	Formaldehyde destruction efficiency of air cleaner E_f	TVOC Si/V (ppb/h)	Formaldehyde Source Strength [No Air Cleaner] S_f/V (ppb/h)	Indoor TVOC Decrease With Air Cleaning	Indoor Formaldehyde Decrease with Air Cleaning
1	2	2	4	5	6	7	8	9	10	11	12	13	14	15	16	17
8	0	0	0.5	46.4	0.32	12.0	0.82	0.26	0.5	20	1	0.7	18.2	8.5	39%	8%
30	0	0	0.5	40.3	0.33	5.6	0.42	0.14	0.5	20	1	0.7	15.2	8.5	40%	35%
8	0	0	0.17	37.2	0.29	7.8	0.73	0.21	0.5	20	1	0.7	13.6	8.5	37%	26%
30	0	0	0.17	41.7	0.46	3.7	0.20	0.09	0.5	20	1	0.7	15.8	8.5	48%	42%
30	0	0	0.5	103	0.23	21.0	0.92	0.20	0.5	20	1	0.7	46.4	8.5	32%	-32%
2.7	5.3	2	0.5?	38.5	0.13	3.8	0.83	0.10	0.5	20	1	0.7	14.2	8.5	21%	41%
4	4	1	0.5?	43.9	0.09	2.5	0.7	0.06	0.5	20	1	0.7	17.0	8.5	15%	47%
5.3	2.7	0.5	0.5?	44.9	0.28	6.2	0.51	0.14	0.5	20	1	0.7	17.4	8.5	36%	32%
10	20	2	0.5?	38.5	0.30	3.5	0.3	0.09	0.5	20	1	0.7	14.2	8.5	38%	44%
15	15	1	0.5?	43.9	0.13	2.9	0.51	0.07	0.5	20	1	0.7	17.0	8.5	21%	46%
20	10	0.5	0.5?	44.9	0.42	3.9	0.21	0.09	0.5	20	1	0.7	17.4	8.5	46%	42%
8	0	0	0.5	46.4	0.32	12.0	0.82	0.26	0.5	40	1	0.7	18.2	18.5	39%	33%
30	0	0	0.5	40.3	0.33	5.6	0.42	0.14	0.5	40	1	0.7	15.2	18.5	40%	47%
8	0	0	0.17	37.2	0.29	7.8	0.73	0.21	0.5	40	1	0.7	13.6	18.5	37%	42%
30	0	0	0.17	41.7	0.46	3.7	0.20	0.09	0.5	40	1	0.7	15.8	18.5	48%	50%
30	0	0	0.5	103	0.23	21.0	0.92	0.20	0.5	40	1	0.7	46.4	18.5	32%	13%
2.7	5.3	2	0.5	38.5	0.13	3.8	0.83	0.10	0.5	40	1	0.7	14.2	18.5	21%	50%
4	4	1	0.5	43.9	0.09	2.5	0.7	0.06	0.5	40	1	0.7	17.0	18.5	15%	53%
5.3	2.7	0.5	0.5	44.9	0.28	6.2	0.51	0.14	0.5	40	1	0.7	17.4	18.5	36%	45%
10	20	2	0.5	38.5	0.30	3.5	0.3	0.09	0.5	40	1	0.7	14.2	18.5	38%	51%
15	15	1	0.5	43.9	0.13	2.9	0.51	0.07	0.5	40	1	0.7	17.0	18.5	21%	52%
20	10	0.5	0.5	44.9	0.42	3.9	0.21	0.09	0.5	40	1	0.7	17.4	18.5	46%	50%
8	0	0	0.5	46.4	0.32	12.0	0.82	0.26	0.5	20	2	0.7	18.2	8.5	56%	10%
30	0	0	0.5	40.3	0.33	5.6	0.42	0.14	0.5	20	2	0.7	15.2	8.5	57%	44%
8	0	0	0.17	37.2	0.29	7.8	0.73	0.21	0.5	20	2	0.7	13.6	8.5	54%	32%
30	0	0	0.17	41.7	0.46	3.7	0.20	0.09	0.5	20	2	0.7	15.8	8.5	65%	53%
30	0	0	0.5	103	0.23	21.0	0.92	0.20	0.5	20	2	0.7	46.4	8.5	48%	-41%
2.7	5.3	2	0.5?	38.5	0.13	3.8	0.83	0.10	0.5	20	2	0.7	14.2	8.5	34%	52%
4	4	1	0.5?	43.9	0.09	2.5	0.7	0.06	0.5	20	2	0.7	17.0	8.5	26%	59%
5.3	2.7	0.5	0.5?	44.9	0.28	6.2	0.51	0.14	0.5	20	2	0.7	17.4	8.5	53%	40%
10	20	2	0.5?	38.5	0.30	3.5	0.3	0.09	0.5	20	2	0.7	14.2	8.5	55%	55%
15	15	1	0.5?	43.9	0.13	2.9	0.51	0.07	0.5	20	2	0.7	17.0	8.5	34%	58%
20	10	0.5	0.5?	44.9	0.42	3.9	0.21	0.09	0.5	20	2	0.7	17.4	8.5	63%	53%

Models inputs not shown include outdoor TVOC concentration of 10 ppb, outdoor formaldehyde concentration of 3 ppb

Table 4 has three sets of rows of model predictions provided in columns 16 and 17. A comparison of the first (un-shaded) and second (shaded) set of model outputs illustrates the modeled effects of assuming two different indoor formaldehyde concentrations prior to application of air cleaning (20 and 40 ppb). A comparison of the first and third set of model predictions (both unshaded) illustrates the predicted effect of assuming two different air cleaner sizes and corresponding air flow rates per indoor air volume (1 and 2 h^{-1}). For completeness, the experimental data are listed three times to correspond with each set of model predictions; however, only one set of 11 experiments was performed.

Consider first the top un-shaded set of rows in table 4. The assumed inlet formaldehyde concentration is 20 ppb, a typical value for homes. The volume-normalized air cleaner flow rate is 1 h^{-1}, a practical value but one that might require 1.5 to two ITAC air cleaners of the size developed for this project for a typical-sized single family home. The modeling indicates a reduction of TVOC concentration ranging from 15% to 48% with air cleaner operation, and in all but one case a simultaneous reduction in indoor formaldehyde concentration ranging from 8% to 47%. The best overall results are a 48% reduction in TVOC and 42% reduction in formaldehyde with 30 g/m^2 of catalyst, no activated carbon, and an air velocity of 0.17 m/s through the catalyst treated filter. Almost identical reductions in both TVOC and formaldehyde concentrations are predicted with 20 g/m^2 of catalyst and 10 g/m^2 of activated carbon on the treated filter and a higher air velocity of 0.5 m/s through the treated filter. However, these predictions are for a moderate to low indoor TVOC concentration of approximately 40 ppb (excluding the concentration of formaldehyde). If the initial TVOC concentration is increased to 103 ppb, even with 30 g/m^2 of catalyst the indoor formaldehyde concentration is predicted to increase by 32%.

For the middle (shaded) set of rows in table 4, the assumed formaldehyde concentration without air cleaning was increased to 40 ppb. In these cases, the modeled TVOC concentrations always decreases (again by 15% to 48%) with air cleaner operation and modeled formaldehyde concentrations also always decrease by 13% to 53%. The best predicted overall performance is an approximate 50% reduction in both formaldehyde and TVOC when the air cleaner is operated with 30 g/m^2 of catalyst.

For the bottom (unshaded) set of rows in Table 4, the assumed indoor formaldehyde concentration without air cleaning is returned to 20 ppb and the air cleaner air flow rate per unit indoor air volume is increased to 2 h^{-1}. The modeling predicts a reduction in TVOC concentration of 26% to 65% with air cleaner operation. Formaldehyde concentrations are predicted to decrease by 10% to 59%, except for the case with the higher 103 ppb of TVOC (excluding formaldehyde) when the air cleaner is not operating. For this case, the model predicts a 41% increase in indoor formaldehyde with air cleaner operation.

3.6 Outreach to facilitate product commercialization

The plans for this research project included outreach to industry with the goal of identifying a company interested in further developing and marketing an air cleaner based on the ITAC design. The outreach effort was scaled back when it became evident from field study data that the ITAC units were not reducing formaldehyde levels as anticipated. However, several air cleaner manufacturers have expressed interest in incorporating filters, treated with LBNL's manganese oxide catalyst, in their products. One company, is actively evaluating catalyst-treated filters, and providing a modest amount of financial support to LBNL to facilitate the evaluations. Another small company is pursuing synthesis of the manganese oxide catalyst via a different method than used by LBNL, for potential incorporation in their air cleaners.

CHAPTER 4:
Discussion

The ITAC units provided or exceeded the target airflow of 71 L/s (150 cfm) at the medium and high fan speed settings with impressively low fan power (approximately 13 W at medium fan speed and 29 W at high fan speed). The goal was a power consumption less than 60 W; thus, the ITAC units exceed this goal by a large margin. Both the ITAC1 and ITAC2 designs were based on easy modifications to an existing energy efficient air cleaner with a known retail price of $460. The incremental cost to modify the original products and mass produce air cleaners with the ITAC1 or ITAC2 design would be modest; thus, the anticipated final costs meet the cost target of $600 for a unit with a 71 L/s supply air flow rate. An alternate design for a more complex ITAC unit incorporating an activated carbon fiber component, regenerated each day, was also developed, but this unit would likely not meet cost targets.

In the short term laboratory studies, formaldehyde removal efficiencies of both ITAC1 and ITAC2 were between 50% and 60% after 24 hours, roughly consistent with expectations based on prior small-scale laboratory tests with sections of catalyst-treated filters (Sidheswaran, Destaillats et al. 2011, Sidheswaran, Destaillats et al. 2011a). In these prior studies, the formaldehyde removal efficiency decreased from approximately 80% to 50% over 100 days. In the current study's short term laboratory tests, VOC removal efficiencies (for which few prior data were available) varied from negligible or slightly negative to approximately 45% for ITAC1 which did not include any activated carbon. The erratic removal efficiencies for hexane, butanal, and benzene in ITAC1 were unexpected, and appear to be only partially explained by the low concentrations of these VOCs during the tests resulting in high measurement uncertainty. The trend toward increased VOC removal efficiency over time in ITAC1 was also unexpected and is unexplained. ITAC1 had been placed in a VOC-laded atmosphere without operation for an extended period prior to the start of tests; thus, it is possible that VOCs adsorbed on the filter elements were being released during the first hours of operation. In the short term laboratory studies, the VOC removal efficiencies of ITAC2, which incorporated activated carbon, ranged

from approximately 25% to 90%. The activated carbon clearly increased the VOC removal, but not formaldehyde removal, in the short term.

In the laboratory studies, the steady state formaldehyde removal efficiencies of ITAC1 and ITAC2 were similar after 24 hours. These findings indicate that the activated carbon in ITAC2 did not contribute substantially to formaldehyde removal. This finding was expected. A review of prior studies of the performance of granular activated carbon has indicated its low capacity for formaldehyde (Fisk 2007).

In the field studies, the formaldehyde removal efficiencies of both ITAC1 and ITAC2 were far less than anticipated based on the prior studies (Sidheswaran, Destaillats et al. 2011, Sidheswaran, Destaillats et al. 2011a) of formaldehyde removal by catalyst treated filters, and were also much less than expected based on data from studies of ITAC performance in the laboratory. In addition, as shown in Figure 11, indoor formaldehyde concentrations were not obviously decreased during periods of ITAC2 operation in H2. Incomplete decomposition of selected VOCs by the catalysts, leading to formaldehyde as a partial decomposition product was the hypothesized explanation for the discrepancy between formaldehyde removal in the field studies and prior data on formaldehyde removal. Another possibility is that formaldehyde adsorbed on the air cleaning media in the ITAC units during periods without air cleaner operation and was released during the subsequent periods of air cleaner operation. This process of adsorption followed by desorption would be more pronounced in the ITAC2 unit containing activated carbon.

Operation of the ITAC units in the two houses appeared to moderately reduce indoor concentrations of VOCs other than formaldehyde. However, given the variability in indoor VOC emission rates and, to a lesser extent, in outdoor air ventilation rates, the evidence that the air cleaner lowered indoor VOC concentrations was not fully conclusive. There was no evidence from the field study data that ITAC2, which contained activated carbon, performed better that ITAC 1 with respect to VOC removal. Thus, the enhanced VOC removal of ITAC2 observed in the laboratory studies may be short-lived. At present, insufficient data are available to determine how long activated carbon filters will remain effective in removing VOCs in complex real-world applications (Fisk 2007).

In H1, indoor particle count concentrations were clearly and substantially decreased during ITAC1 operation. In H2, there were periods with very high indoor particle concentrations indicating periods of very high indoor particle emission rates. Ignoring these periods, the data indicate a moderate roughly 50% decrease in indoor particle concentrations during ITAC operation.

Because of limitations in instrumentation, it was not possible to perform air pollutant measurements at multiple indoor locations. VOC concentrations were measured at the inlet and outlet of the air cleaners and particle concentrations were measured at the inlet of the air cleaners. These measurement locations enabled a calculation of the VOC removal efficiency of

the air cleaners. In general, the doors to the rooms containing air cleaners were maintained open to facilitate mixing of air with other rooms in the house. However, due to imperfect mixing the air cleaners likely changed the average pollutant concentrations in the full homes less than indicated by measurements made at the inlets of the air cleaners.

The final laboratory studies confirmed that formaldehyde can be produced by incomplete decomposition of VOCs by the catalyst-treated filters. Thus, incomplete VOC decomposition and associated formaldehyde production may explain why the formaldehyde removal efficiencies in the field studies were much less than expected. This issue may have been less evident in the laboratory studies because of the different mixture of VOCs in laboratory air and house air. The results shown in Table 3 suggest that formaldehyde can be produced as a byproduct of incomplete decomposition of many VOCs. The higher yield of formaldehyde for styrene is consistent with the presence of a reactive vinyl functional group that can readily produce formaldehyde in the first oxidation step. The presence of unusually high styrene levels in the air of H1 may explain the limited formaldehyde removal efficiency during testing in this house. A similar high formaldehyde yield can be expected from incomplete decomposition of terpenoids, such as d-limonene, commonly used in cleaning products. The problem of incomplete VOC decomposition and formaldehyde production is not unique to this air cleaner design. Ultra-violet photo-catalytic oxidation air cleaning systems share this problem (Hodgson, Destaillats et al. 2007) and many ultra-violet photo-catalytic oxidation air cleaners are available as commercial products. Improving the engineering of photo-catalytic oxidation air cleaners by incorporating more catalyst and increasing the residence time for VOC contact with the catalyst has improved the VOC and formaldehyde removal efficiency, resulting in a net formaldehyde removal in some cases (Destaillats, Sleiman et al. 2012).

In this study, with mixtures of VOCs but no formaldehyde entering sections of catalyst-treated filters, the amount of formaldehyde produced per unit of VOC decomposition was substantially diminished by increasing the amount of catalyst on the filter and also by decreasing the air velocity. Together, these two measures reduced formaldehyde yield (formaldehyde production, per unit VOC removal) by a factor of four, while increasing the VOC removal efficiency by a factor of 1.4. Limited testing indicated that substituting powdered activated carbon for a portion of the catalyst had a modest effect on VOC removal efficiency and formaldehyde yield; however, this measure may be attractive because the powdered activated carbon will cost less than the catalyst.

Laboratory tests and modeling results suggest that a modified ITAC1 air cleaner, with a larger amount of catalyst (10 g/m^2 or higher) or with at least 10 g/m^2 catalyst plus additional powdered activated carbon, and with a lower air velocity through the catalyst treated filter, will simultaneously reduce indoor formaldehyde concentrations and concentrations of other VOCs when initial concentrations of VOCs other than formaldehyde are low or moderate. These changes could be implemented by applying the catalyst, or the catalyst plus activated carbon, to a pleated filter with a larger surface area than the filter to which catalyst was applied in ITAC 1. One option would be to apply the catalyst to the highly pleated HEPA filter of ITAC1; however,

this would shorten the interval between replacements of this expensive filter. For this reason, applying catalyst to a less expensive pleated medium-efficiency filter might be preferable. Alternately, the pleated catalyst-treated filters in ITAC air cleaners might be replaced with catalyst treated pad filters that have a thicker filter media. In the small scale laboratory studies, catalyst has been applied to a 2.5 or 5 cm thick pad filters, while the filter media in the pleated filters is much thinner, on the order of 1 mm. Model results suggest that increasing the amount of catalyst and decreasing the air velocity are not sufficient measures for homes with initially high concentrations of VOCs other than formaldehyde (i.e., strong indoor VOC sources), as formaldehyde concentrations may be increased during air cleaner use.

While the results of studies of methods to reduce formaldehyde production from incomplete VOC decomposition were very encouraging, the results of the modeling employed to facilitate interpretation of these results should be treated with much caution. The experimental data underlying the model were too limited to enable development of a validated model. In these experiments, there was no formaldehyde in the inlet airstream. Omitting formaldehyde from the air entering the catalyst-treated filters enabled measurements of formaldehyde production rates from incomplete VOC decomposition; however, the air entering air cleaners from real buildings will contain formaldehyde. The formaldehyde removal efficiency used in modeling was based on the results of prior experiments with only formaldehyde in the air entering catalyst treated filters. The mixture of VOCs employed in the experiments contained fewer VOCs, with significant concentrations, than the air in most buildings, and the composition and concentration of VOCs will influence the rates of formaldehyde production from incomplete VOC decomposition. Additional, experiments, including field trials, are needed to determine how well a modified air cleaner will work in practice.

CHAPTER 5:
Conclusions

The project was able to design and test two air cleaners for simultaneously removing particles and VOCs in residential applications. The designs met the target for air cleaner flow rate, maximum air cleaner energy use, and air cleaner cost. Thus, if the air cleaners had met all goals with respect to particle and VOC removal, they would have had the potential to maintain indoor air quality at a lower energy cost than outdoor air ventilation. The project also produced an alternative design (see appendix) for a more complex air cleaner, employing periodically-regenerated activated carbon fiber cloths for VOC removal; however, this product is likely to be too costly for widespread use.

While the air cleaners removed formaldehyde as expected in short term laboratory studies, their formaldehyde removal performance was much less effective than anticipated during air cleaner operation in houses.

The laboratory study data showed that the catalyst-treated filters were moderately effective in removing a range of VOCs other than formaldehyde. Few prior data were available on removal of VOCs other than formaldehyde. Data from field studies also indicated VOC removal, but were not conclusive due to considerable temporal variability in indoor VOC emission rates.

The field study data also indicated that the air cleaners reduced indoor particle concentrations, particularly the concentrations of small particles. These findings are consistent with prior data showing that similar air cleaners with HEPA filters are effective in reducing indoor particle concentrations in homes.

Controlled studies in the laboratory, following the field studies, confirmed that the catalyst-treated filters, similar to the filters used in ITAC1 and ITAC2, can produce formaldehyde via incomplete decomposition of some VOCs.

Controlled laboratory studies showed that the amount of formaldehyde produced from incomplete VOC decomposition, per unit VOC destroyed, can be decreased by a factor of four by increasing the amount of catalysts applied to the filter and decreasing the air velocity.

Modeling suggests that a modified version of ITAC1, with catalyst applied to a filter with a larger surface area, will be effective in simultaneously reducing formaldehyde concentrations and concentrations of other VOCs in homes when initial concentrations of VOCs (other than formaldehyde) are low and/or initial concentrations of formaldehyde are high. However, further experiments are necessary to determine how well this design will work in practice.

GLOSSARY

| Term	Definition
ACF	Activated carbon fiber
DNPH	Dinitrophenyl hydrazine
H1	House number 1
H1	House number 2
ITAC	Integrated technology air cleaner
HEPA	High efficiency particle air
ITAC1	Integrated technology air cleaner number 1
ITAC2	Integrated technology air cleaner number 2
MERV	Minimum efficiency reporting value
TVOC	Total volatile organic compound
VOC	Volatile organic compound

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APPENDIX A:
Alternate ITAC Design

This appendix describes an alternate design for a more complex ITAC air cleaner that incorporates, in addition to the particle filter and catalyst-treated filter, an activated carbon fiber (ACF) element that is regenerated in-situ with heated air. The performance and operational specifications for ACF air cleaning have been described in a prior document [A1]. When an ACF element is included, there is a need to be able to periodically stop the flow of indoor air through the air cleaner, isolate the ACF filter element from indoor air, bring in outdoor air for regeneration, heat this air to 150 °C, pass the heated air through the ACF media, and vent the VOCs desorbed from the ACF to outdoors. Sections of small-diameter tubes connecting the air cleaner to outdoors are necessary. One tube brings outdoor air to the air cleaner for use in the regeneration of the ACF media and the second tube vents this air plus VOCs desorbed from the ACF to outdoors. For a 71 L/s (150 cfm) air cleaner, a regeneration airflow rate of 0.7 L/s (1.5 cfm) is adequate. Even lower regeneration airflow rates, or lower regeneration air temperatures and longer regeneration times may be effective, but these alternatives would need to be evaluated experimentally. With no heat recovery between the incoming and outgoing regeneration air, and no conductive losses, a 100W heater, operated 15 minutes per day, is needed for the 150 °C regeneration temperature. The apparatus holding the ACF cloth must be able to withstand 150 °C temperatures.

An obvious potential configuration is shown in Figure A1, with electrically actuated dampers located upstream and downstream of the ACF element. These dampers must close and isolate the ACF media from the indoor air during the regeneration cycle and be able to withstand 150 °C temperatures. While such a damper system is very feasible technically and likely practical in larger commercial building applications, practical-cost dampers and actuators for a small residential air cleaner were not identified.

Figure A1. Initial concept for ITAC backup design.

(● = heating element for regeneration)
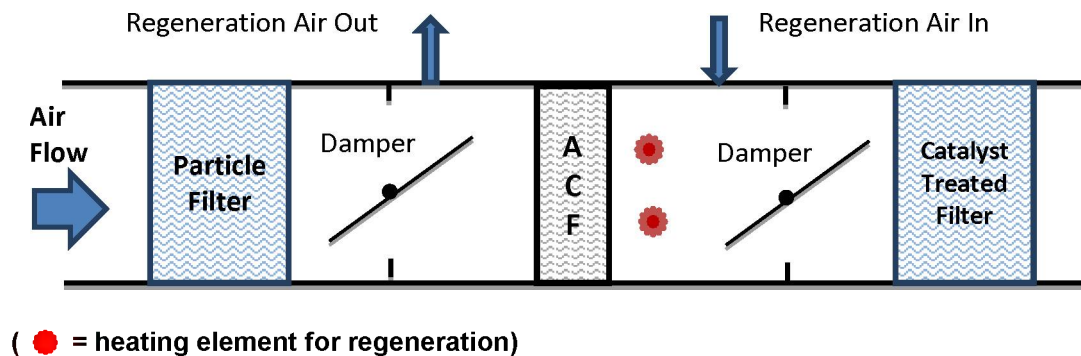
Given the high cost of damper actuators and dampers able to withstand 150 °C temperatures, the current leading design incorporates a moving ACF filter box. While non-conventional, with this design only one screw-type or linear actuator is required, seals that withstand high temperatures and isolate the ACF unit from indoor air during regeneration cycles are straight forward, and the heated regeneration section is well isolated from the remainder of the air cleaner and more easily thermally insulated. Figure A2 shows a cross section of the system configuration, with the ACF filter element shown in the position for air cleaning on the left and the ACF filter element in the position for regeneration on the right. Not shown in this schematic is a simple heat exchanger for preheating the incoming regeneration air with the outgoing regeneration air, the access panels for replacing filters, controls, and a few other elements. In another version of this design, requiring evaluation, the connections to outdoors are eliminated and VOCs in the heated regeneration air stream are destroyed by passing this airstream through a bed of catalyst. This alternate ITAC design could employ the HEPA filter and high efficiency fan system from the ITAC1 and ITAC2 designs, or equivalent products. The HEPA filter is expected to cost approximately $50. A lower cost filter with a MERV 14 efficiency rating could be substituted for the HEPA filter. The heating elements needed for regeneration can be simple light bulbs used in residential ovens. The few seals required are made of Viton, which withstands high temperatures and is not a significant pollutant source. The air cleaner cross sectional dimensions would be approximately 0.46 m by 0.46 m (1.5 ft by 1.5 ft). The filter treated with catalyst can be a fiberglass or synthetic pad filter from American Air Filter (used successfully as a substrate for the catalyst in past experiments) or a low efficiency 2.5 cm (1 inch) thick pleated filter.

In Figure A2, the catalyst-treated filter is shown downstream of the ACF element but the catalyst treated filter could be placed upstream of the ACF element. Also, if a double layer of the ACF is utilized, and the velocity through the ACF cloth is maintained at 0.5 m/s or less, a formaldehyde removal efficiency of 40% could be maintained while eliminating catalyst treated filter. This option would eliminate the risks associated with incomplete decomposition of VOCs by the catalyst.

The moving and heat-resistant ACF filter system is the most complex element of the design. Figure A3 shows one potential configuration for this unit with ACF filter sections that slide into slots of a metal filter box. Not shown are the drawer slides at each end of the filter box that support the box and guide its movement. These drawer slides, sometimes used in file cabinets, employ rows of ball bearings in track systems to ease and guide the linear back-and-forth movement of a drawer. The ACF filter sections shown in Figure A4, are simple sections of ACF cloth, sandwiched between upper and lower perimeter metal frames, with the upper and lower sections of frame held together by spot welds.

References to Appendix A

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FigureA2. Cross section schematic of the backup ITAC system configuration with the ACF filter element shown in the position for air cleaning on the left and the ACF filter element in the position for regeneration on the right.
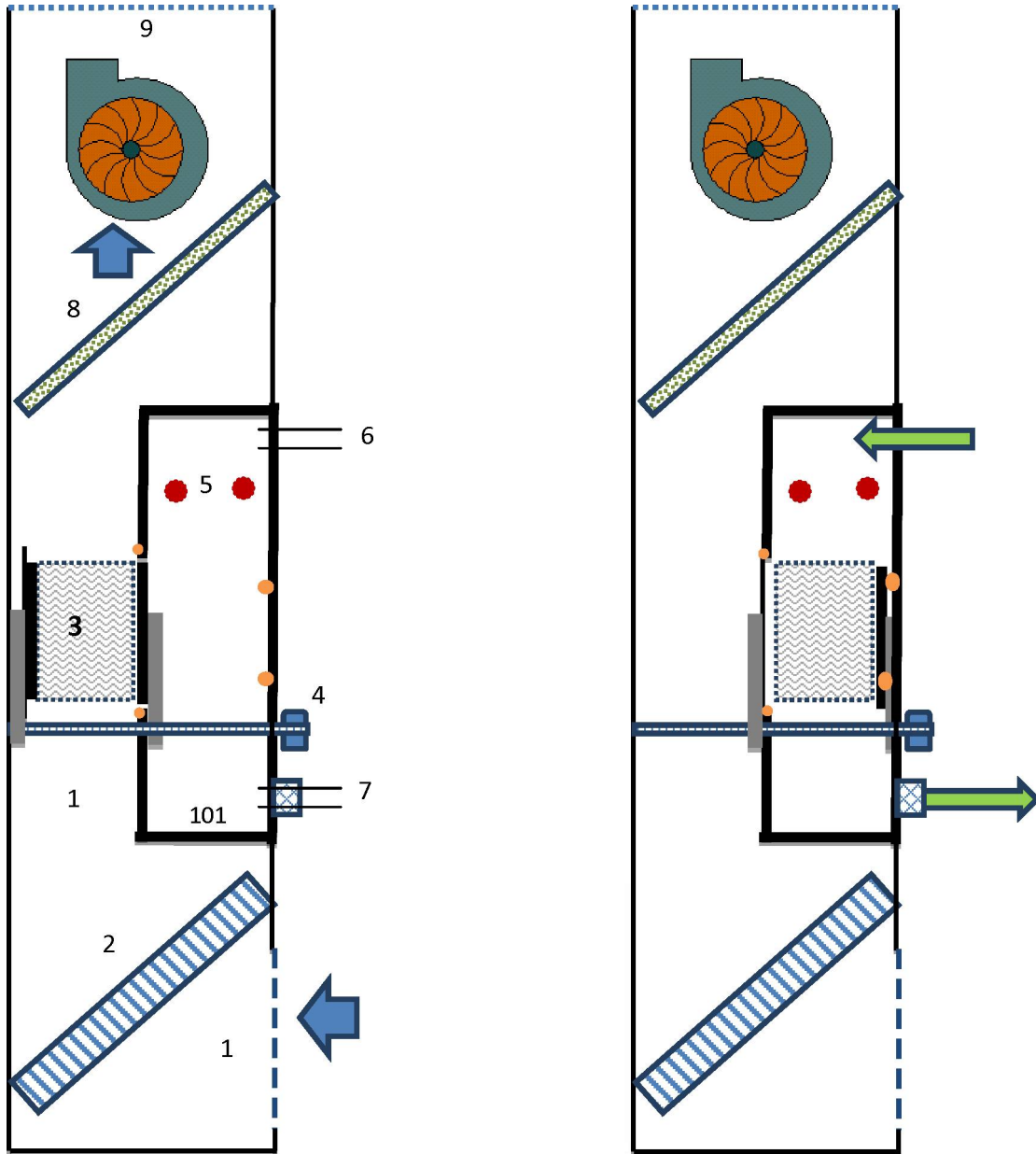
Key: 1 = inlet screen   2 = high efficiency particle filter   3 = ACF filter unit   4 = screw drive and motor actuator for ACF unit   5 = regeneration air heater lamps   6 = regeneration air inlet 7 = regeneration air outlet and fan   8 = catalyst treated filter 9 = fan   10 = double-wall insulated perimeter of regeneration air heated chamber

Figure A3. Schematic cross section view of the ACF filter box.

Side plate

Slot in side plate, holds edge of ACF filter element
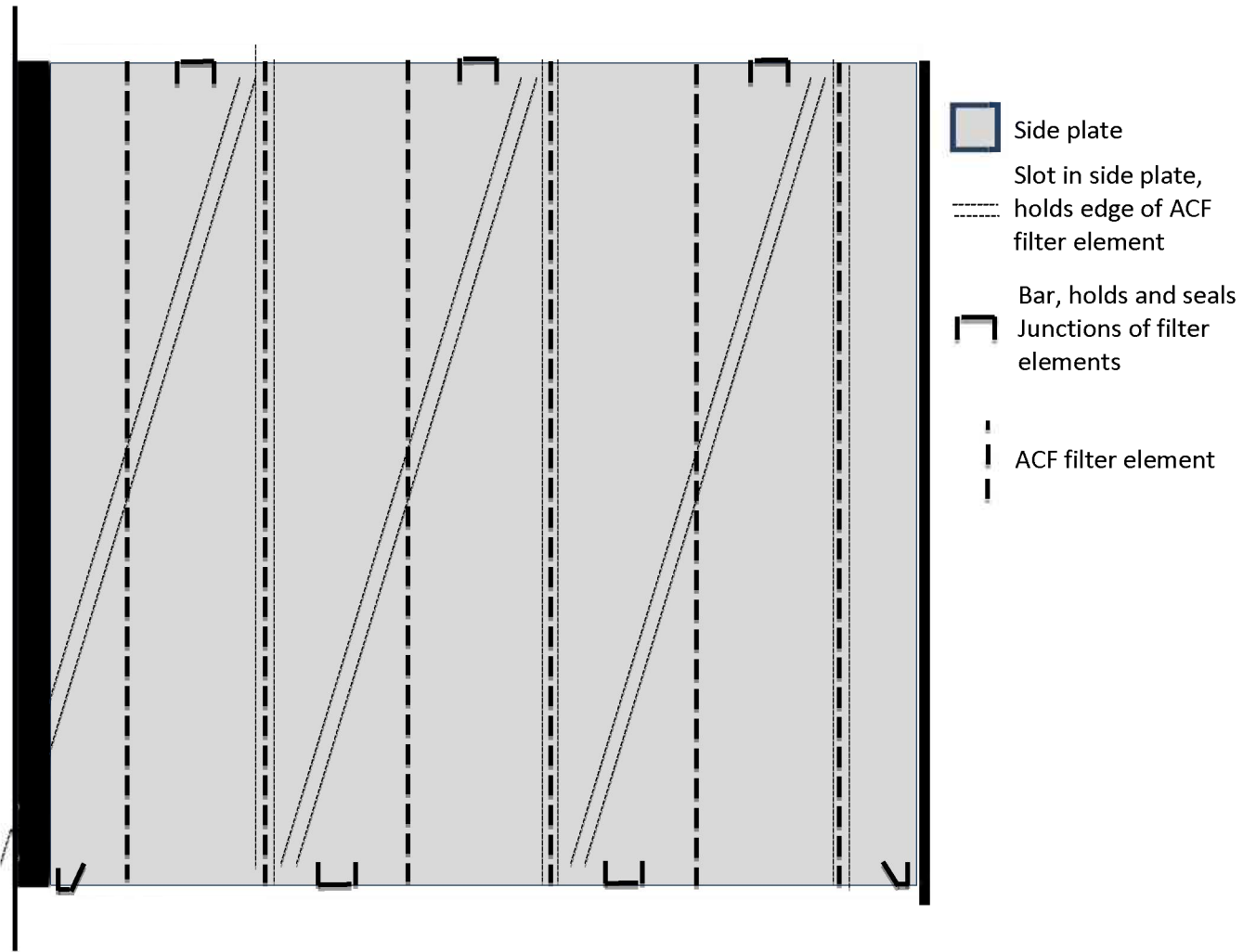
Bar, holds and seals Junctions of filter elements

ACF filter element

Figure A4. ACF filter section

ACF filter
section

ACF
Cloth

Metal edge
plate

Side view showing
metal plates at
edges of ACF filter
cloth. Plates are
held together with
spot welds.