



# Exposure to cooking emitted volatile organic compounds with recirculating and extracting ventilation solutions

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## ABSTRACT

Energy-efficient urban development leads to the compact design of apartments. Recirculating ventilation solutions are an attempt to minimize the space required for ventilation ducting, but more data on their performance are needed. Cooking is a major source of volatile organic compounds (VOCs) emissions. It is necessary to assess how well recirculating kitchen hoods perform in reducing the residents' exposure to cooking fumes compared to extracting hoods, and what airflow rates assure good removal efficiency. We have monitored the occupant exposure to several VOCs generated during the cooking of a model meal under different ventilation scenarios in a purpose-built test kitchen resembling the layout of a modern, open-space apartment. Time-resolved VOC emission profiles were measured using a proton transfer reaction time-of-flight mass spectrometer. The performance of activated carbon sorption-based filters for recirculating kitchen hoods in the removal of selected VOCs was also assessed. Alcohols, particularly ethanol, dominated emissions from cooking a typical Norwegian meal, but they also included acetaldehyde, acetone, carboxylic acids, and trimethylamine, among others. The use of recirculating kitchen hoods led to, on average, higher occupant exposure to VOCs compared to extracting kitchen hoods during and after cooking. This was in part due to the poor ethanol removal efficiency of the recirculating ventilation's air filters.

## 1. Introduction

Resilient and sustainable urban development in times of increased urbanisation [1] creates demand for compact apartments in downtown areas and near junction points. Thus, apartment buildings are becoming taller, and the apartments within less spacious, with space-efficient open-plan layouts. Exposure to noise and outdoor atmospheric pollution near high-traffic areas limits the possibility of airing the apartment by opening the windows, thus increasing the ventilation system's role in removing indoor pollutants. This is compounded by the trend towards energy-efficient buildings and neighbourhoods in the EU [2] and necessitates balanced ventilation systems with effective heat recovery.

Since cooking is a major source of indoor air pollution [3,4], effective kitchen ventilation plays a crucial role in ensuring a healthy indoor environment and factors into the energy budget. Extracting hoods are the most effective solution for removing cooking emissions [5], provided their capture efficiency is sufficient, which might require high airflow

rates. This, however, creates new challenges: under-pressure in airtight buildings, regulation of makeup air, and heat recovery. In the particular context of compact urban apartments, there is also the problem of the availability of space for ductwork. These issues have given rise to alternative solutions, *i.e.* recirculating kitchen ventilation systems which remove cooking emissions using an air filter and the purified air is returned to the room. This eliminates the need for makeup air and does not require a connection to the balanced ventilation system. However, compared to the established solutions using extracting hoods, there is a lack of reliable evidence for the effectiveness of the recirculating systems in reducing the occupant's exposure to cooking emissions [6]. Moreover, studies have shown that higher airflow rates than the ones prescribed by the building codes (e.g.  $108 \text{ m}^3 \text{ h}^{-1}$  in the case of Norway at the time of writing) might be needed to ensure good capture efficiency and indoor air quality [5,7,8].

The Norwegian research project "Healthy Energy-efficient Urban Home Ventilation" addresses ventilation solutions and related

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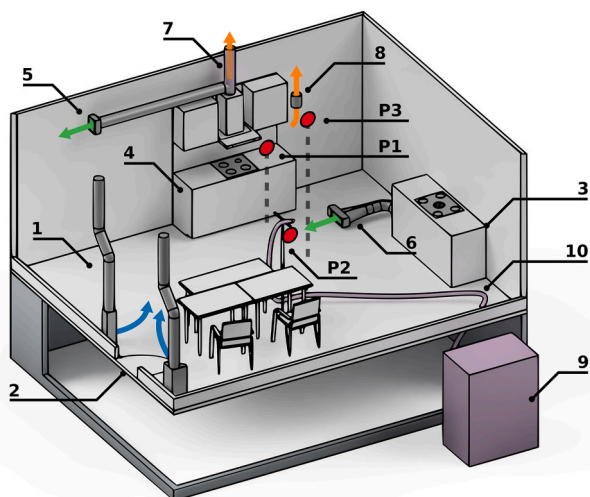
challenges for highly energy- and space-efficient urban dwellings. Part of the study involved investigating the effect of kitchen ventilation solutions and different ventilation rates on the occupant's exposure to cooking emissions, both particles and volatile organic compounds (VOCs) in a compact, open-space apartment. This was done in a purpose-built test kitchen.

We intend to present the results focusing on the emission of cooking aerosol, including fine and ultra-fine particles, in a separate paper. Here, we focus on the results related to the emission of VOCs in the gaseous phase, which was monitored using a proton-transfer-reaction time-of-flight mass spectrometer (PTR-ToF-MS). The aim here was to assess the performance of recirculating kitchen hoods versus extracting kitchen hoods in reducing the resident's exposure to VOCs, including odorants, at different ventilation rates. We also investigated the efficiency of activated carbon sorption-based filters in removing volatile organic compounds generated during cooking, having first characterized the VOCs emission profile of cooking a typical Norwegian meal.

## 2. Materials and methods

### 2.1. Test kitchen setup

The cooking emission experiments were carried out in a purpose-built test kitchen with a footprint of 480 cm × 620 cm and a height of 270 cm, situated within a larger laboratory (see Fig. 1). A detailed description of the test kitchen and its ventilation setup was provided elsewhere [8]. The room emulates an open-plan apartment and is thus larger than the test room described in EN 13141-3 [9] and IEC 61591. While similar designs exist for simulating the ventilation in a separate kitchen [10], or in a separate kitchen with an adjacent living room [11], such arrangements are becoming less common in European apartments. The room featured a single doorway sealed with a rubber gasket when closed. A ceiling-mounted air terminal coupled to an exhaust fan was used to maintain a baseline ventilation rate of 36 m<sup>3</sup>h<sup>-1</sup>, *i.e.* the minimum for kitchen ventilation according to Norwegian regulations [12]. A mechanical ventilation system supplied the lab with well-filtered air through two displacement diffusers placed on either side of the



**Fig. 1.** The test kitchen experimental setup. The room featured a clean air supply through displacement diffusers (1) on both sides of the door (2) and two cooking stations equipped with either a downdraft (3) or standard (4) ventilation setup. The kitchen hoods were operated in recirculating (5, 6) and extracting (7) modes. The main air extract was through a ceiling-mounted vent (8). The cooking emissions were sampled within the room and transferred to a PTR-ToF-MS device placed outside (9) via a heated transfer line (10). The VOCs were sampled at 3 locations: next to the cook's position (P1), in the middle of the room (P2), and at the main air extract (P3).

entrance. The supply rate was adjusted to maintain a 0.5 Pa ± 0.2 Pa overpressure within the test kitchen. This was done to eliminate the risk of VOCs from outside the room interfering with the measurements. With the additional flow necessary to maintain the overpressure the overall air change rate at baseline ventilation was 0.76 ± 0.07 h<sup>-1</sup> (mean ± std, n = 3) based on a gas tracer measurement. During the operation of the extracting kitchen hood setups, the air supply was increased to match the combined ventilation rate. Pressure, relative humidity, and temperature were monitored throughout the experiments.

Two induction cooktops were placed along perpendicular walls, equidistant from the ceiling-mounted extract. A wall-mounted kitchen hood (henceforth referred to as "standard") which can be operated at both exhaust and recirculation mode was mounted 54 cm above the cooktop situated along the longer wall. When configured for recirculation, the fumes were directed away from the cooking area through a 220 cm segment of a ventilation duct to allow for reliable flow control and regulation, before being passed through an activated carbon filter. A similar arrangement was used for the downdraft setup, with the airflow passed through a duct along the floor and an activated carbon filter back into the test kitchen. For reference, the objects in the isometric room projection in Fig. 1 are true to scale. The flow in both the extracting and the recirculating mode was measured using the 3000MD micro-manometer (Swema AB, Sweden) connected to an in-line differential pressure measuring station (BAAS Component AS, Norway) situated no less than five times the duct diameter (5 × ø160 mm) from the hood's fan.

### 2.2. Cooking procedure

All operations within the test kitchen were performed by a single researcher who remained in the room for the duration of the experiment. The model meal that was cooked throughout the measurement campaign was selected based on a survey that revealed typical Norwegian home cooking habits [13]. It was composed to produce a balanced and nutritious dinner suitable for two adults and consisted of fried fish, rice, and vegetables. The development of the cooking procedure was described elsewhere [13]. Briefly, the meal preparation involved frying 500 g of salmon fillets, without skin, packaged in modified atmosphere packaging, followed by frying a pre-cooked, deep-frozen rice and vegetables stir-fry mix (500 g). The products were obtained from a local retailer in Oslo, Norway, transferred to the laboratory within 10 min of purchase, and refrigerated until the experiment. Both the fish and the stir-fry mix were fried in non-stick frying pans with 5 mL of rapeseed oil. The cooktops were set to a setting that produced pan surface temperatures closest to 170 °C for frying salmon and 100 °C for frying the stir-fry mix. The salmon fillets were seasoned with salt and black pepper before frying.

The first 5 min of each experiment were dedicated to background measurement. In scenarios in which the extracting setup was used, the overall ventilation rate was increased 30 s before the kitchen hood was turned on to make up for the extracted air and returned to the base value 30 s after it was turned off. If the kitchen hood was used, it was turned on 5 min after the beginning of the experiment ( $t_0$ ), together with the first burner. Salmon was fried from  $t_0 + 00:06:00$  till  $t_0 + 00:14:30$ , followed by the stir-fry mix from  $t_0 + 00:16:00$  till  $t_0 + 00:21:00$ . At that point the fried products were plated on disposable plates and transferred to the table in the middle of the test kitchen to emulate a scenario in which the occupant sits down to eat the prepared meal. The kitchen hood and stovetop were turned off, and the researcher remained seated at the table until the end of the experiment, *i.e.* until  $t_0 + 01:13:00$ . In some experiments the cooked food was not transferred to the table but was instead sealed in an airtight container and left in the cooking area to differentiate between the cooking emissions and the emissions from the cooked meal. A Gantt chart illustrating the cooking procedure is shown in Fig. 2. After each experiment, the stovetops, pans, and cooking utensils were cleaned with dishwashing detergent, and the test kitchen

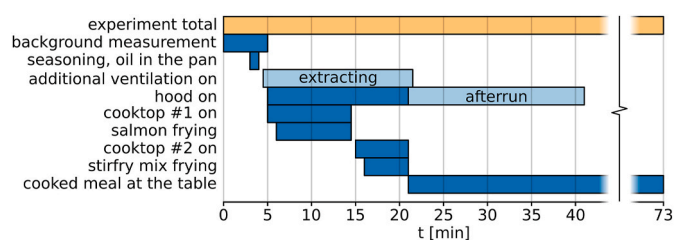


Fig. 2. Gantt chart of the cooking procedure. In some experiments, the cooked meal was not transferred to the table in the middle of the test kitchen and was instead sealed in airtight containers.

was ventilated until the monitored signals returned to baseline and reached a steady state.

During the majority of experiments, the VOCs were sampled in the middle of the room (Fig. 1 P2) 140 cm above the floor level. That was also where the temperature and RH were monitored. The mean temperature throughout the campaign was  $22.5\text{ }^{\circ}\text{C} \pm 0.8\text{ }^{\circ}\text{C}$  and the mean RH was  $20.0\text{ \%} \pm 5.6\text{ p. p.}$  Additional sampling points for the standard extracting (SE) setup were located next to the cook's position 140 cm above the floor level and next to the main ceiling exhaust (Fig. 1, P1 and P3, respectively). Most experiments were focused on assessing the exposure (i.e., the time-averaged VOCs mixing ratio at the point of sampling) of the 'occupant' as opposed to the 'cook', with the volatiles sampled at P2. The hoods' airflow was adjusted to either  $180\text{ m}^3\text{h}^{-1}$  or  $250\text{ m}^3\text{h}^{-1}$ . The former rate was based on preliminary results of capture efficiency of cooking aerosol, and the latter is the common recommendation in Norway [14]. Both hoods were operated with and without a 20-min afterrun of  $80\text{ m}^3\text{h}^{-1}$  when in the recirculation configuration. This is a feature in which the hood remains on, operating at a reduced flow rate for a period after the end of cooking. An overview of the conducted measurements can be found in Table 1. All 21 scenarios were tested in triplicate, with a total of 63 tests. Additional measurements were carried out to assess the contribution to the measured VOCs emission from the researcher (e.g. through exhaled acetone [15]) who followed the motions of the cooking procedure without the actual cooking.

Table 1

An overview of the conducted experiments. The values in the ventilation rate (Q) column indicate the base ventilation rate of the test kitchen, followed by the additional ventilation rate during the hood operation (either extracting or recirculating), and the value in brackets indicates the additional ventilation rate during afterrun.

Designation	Sampling point	Setup	Mode	Q [ $\text{m}^3\text{h}^{-1}$ ]	Afterrun	Dish sealed
P1_S0	P1	Standard	n.a.	36	n.a.	no
P1_SE180	P1	Standard	Extracting	36 + 180	no	no
P1_SE250	P1	Standard	Extracting	36 + 250	no	no
P2_S0	P2	Standard	n.a.	36	n.a.	no
P2_S0S	P2	Standard	n.a.	36	n.a.	yes
P2_SE180	P2	Standard	Extracting	36 + 180	no	no
P2_SE250	P2	Standard	Extracting	36 + 250	no	no
P2_SR180	P2	Standard	Recirculating	36 + 180	no	no
P2_SR250	P2	Standard	Recirculating	36 + 250	no	no
P2_SR180_A	P2	Standard	Recirculating	36 + 180 (80)	yes	no
P2_SR250_A	P2	Standard	Recirculating	36 + 250 (80)	yes	no
P2_D0	P2	Downdraft	n.a.	36	n.a.	no
P2_D0S	P2	Downdraft	n.a.	36	n.a.	yes
P2_DR250	P2	Downdraft	Recirculating	36 + 250	no	no
P2_DR180_A	P2	Downdraft	Recirculating	36 + 180 (80)	yes	no
P2_DR250_A	P2	Downdraft	Recirculating	36 + 250 (80)	yes	no
P2_DR250S	P2	Downdraft	Recirculating	36 + 250	no	yes
P2_DR250S_A	P2	Downdraft	Recirculating	36 + 250 (80)	yes	no
P3_S0	P3	Standard	n.a.	36	n.a.	no
P3_SE180	P3	Standard	Extracting	36 + 180	no	no
P3_SE250	P3	Standard	Extracting	36 + 250	no	no

\* One additional measurement, P2\_SE250S, was discarded during data processing due to very high standard deviation ( $\sigma > 2\bar{x}$ ) and inconsistency of the emission profile with all other measurements.

### 2.3. PTR-ToF-MS measurements

The cooking-related emissions in the test kitchen were monitored using a proton-transfer-reaction time-of-flight mass spectrometer. This technique is widely used in applications in which the measurement of dynamic emission profiles of particular VOCs in the indoor environment is needed [16]. The instrument used in this study was custom-built and was described in detail by Müller et al. [17]. It enables real-time measurement of most VOCs with high temporal (1 Hz) and mass resolution ( $m/\Delta m \approx 2500$ ). The reaction chamber of the instrument was operated at  $75\text{ }^{\circ}\text{C}$ , 3.7 mbar, and 600 V. These settings resulted in a reduced electric field ( $E/N$ ) strength of 97 Td, where  $1\text{ Td} = 10^{-21}\text{ V m}^{-2}$ . The PTR-ToF-MS was operated in the  $\text{H}_3\text{O}^+$  mode. The instrument was placed outside of the test kitchen (see Fig. 1.9). The volatiles were sampled from within the room via a 10 m perfluoroalkoxy alkane (PFA) line heated to  $60\text{ }^{\circ}\text{C}$ , with a sampling flow of  $1\text{ L min}^{-1}$ . The inlet of the line was fitted with a PFA housing for a  $0.45\text{ }\mu\text{m}$  polytetrafluoroethylene (PTFE) filter that was changed periodically (approx. once every 9 experiments). A fraction of the main flow was subsampled into the PTR-ToF-MS analyser.

The instrument was initially calibrated using a multi-component gas calibration mixture containing 15 VOCs (Apel-Riemer Environmental, USA) between  $\approx 10$  and  $0.5\text{ ppm}$  ( $\pm 5\text{ \%}$ ). After the monitored signals were assigned to particular compounds, the instrument was calibrated for these compounds using both dynamically diluted gas standards and volumetrically and gravimetrically prepared aqueous standard solutions. Liquid standards were sourced from Sigma-Aldrich (Norway). The standard solutions were introduced into the instrument via a liquid calibration unit (LCU, Ionicon Analytik, Austria) in which they were nebulized and evaporated in a  $1\text{ L min}^{-1}$  flow of zero air. The LCU was also used to dynamically admix HPLC-grade water into the generated gaseous calibration mixture to account for the change of sensitivity induced by RH changes throughout the experiments. In the case of signals which were not assigned to particular chemical compounds, the sensitivity to acetone was used as a proxy.

The obtained mass spectra ranged from  $m/z$  14 to  $m/z$  350 and were integrated every 5 s. They were then processed using the PTR-MS Viewer 3.4 software (Ionicon Analytik, Austria). The concentration measured

during the first 5 min of each experiment, *i.e.* before the kitchen hoods were turned on and the cooking had begun, was treated as baseline, with the exception of the signal subsequently assigned to monoterpenes, in which case the concentration during the first 2 min of the experiment was treated as baseline (see Fig. S5 for explanation). Only signals with a signal/noise ratio >10 were selected. The list was further curtailed to only include signals that increased in intensity by a factor of at least 1.3 during and immediately after cooking.

PTR-ToF-MS does not provide structural information, making it impossible to discriminate structural isomers. The compounds were assigned to particular ions based on their molecular formulae, relevant literature references [18,19], and whether or not it would be sensible to expect a given compound to be a primary emission from the cooked foodstuffs. Ions that were almost certainly fragments of already assigned compounds based on correlation analysis (e.g., the monoterpenes fragment at  $m/z$  81) were excluded from the list, while ions that could be possibly assigned to several compounds were denoted with their molecular formulae.

The occupant's exposure to a given volatile, *i.e.* its mean mixing ratio at the sampling point, was calculated as the time-weighted average (TWA) of the total emission of the VOCs from the moment the cooktop was turned on at  $t_0+5$  min until the end of the experiment at  $t_0+73$  min:

$$TWA = \frac{1}{n} \sum_{t=5}^{73} c_t,$$

where  $n$  is the number of individual mass spectra recorded from  $t_0 + 5$  until the end of the experiment. In this case, the concentration was averaged over a period of 68 min. The total concentration of VOCs (TVOC) was calculated as the sum of the mixing ratio of the monitored ions (14 in total). The maximum concentration levels of the VOCs were compared with their respective irritation levels and with odour sensitivity thresholds [20,21]. We have focused on assessing the exposure of the residents to cooking emissions, and so calculated the exposure as a time-weighted average over a period of time extending beyond the cooking itself and beyond the period in which the kitchen hoods were operated. This produced less stark differences between the measured efficiency of the ventilation interventions, but overall more representative results.

The capture efficiency (CE) of the kitchen hood operated in the extracting mode was estimated by comparing the TVOC concentration in the middle of the room in scenarios in which cooking took place with ( $C_V$ ) and without ( $C_0$ ) the hood operating. The estimation was based on cross-calculation of each of the triplicate measurements in the different scenarios:

$$CE = \frac{1}{m \times n} \sum_{i=1}^m \sum_{j=1}^n \left( 1 - \frac{C_{Vj}}{C_{0i}} \right) \times 100\%,$$

where  $m$  and  $n$  are the number of  $C_0$  and  $C_V$  measurements, respectively, and  $i$  and  $j$  are the indices of particular replicates in each of the two scenarios. In this case  $m = n = 3$ , so the overall CE was calculated as the mean of 9 combinations.

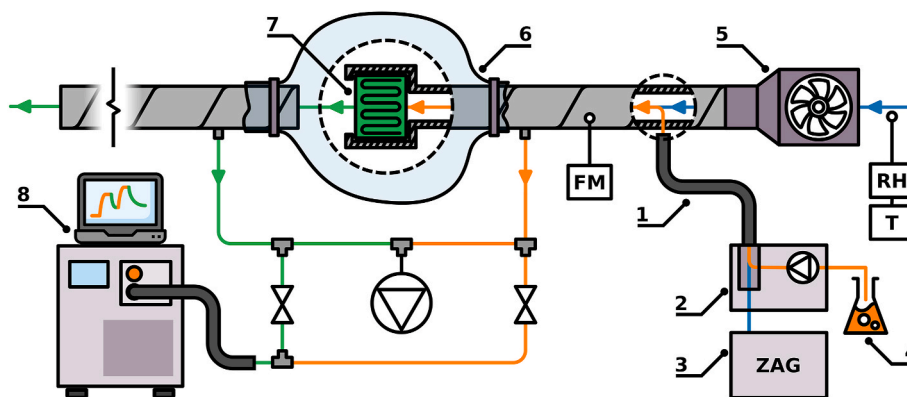
#### 2.4. Filter initial removal efficiency measurements

A separate laboratory setup (Fig. 3) was created to assess the initial removal efficiency ( $E_I$ ) of the filters used in the test kitchen study. Airflows matching the ones used in the recirculating setups during cooking were generated using an in-line ventilation fan connected to a standard  $\varnothing 160$  mm ventilation duct. The airflow was adjusted by varying the fan speed and controlled using the Swema 3000md micromanometer connected to an in-line differential pressure measuring station (BAAS Component, Sweden) positioned 800 mm downstream from the fan. A temperature and RH probe was placed next to the fan's inlet and connected to a continuous data logger. Both filters were mounted downstream from the fan in dedicated housings, that were in turn enclosed in a chemically inert PFA sleeve (Welch Fluorocarbon, USA). A second duct segment channelled the flow downstream of the filter into a laboratory fume hood.

Analytical standards of five VOCs: acetaldehyde, ethanol, trimethylamine (TMA), pinene, and acetic acid were obtained from Sigma-Aldrich (Norway). They were introduced into the LCU (Ionicon Analytik, Austria) at  $0.01 \text{ cm}^3 \text{ min}^{-1}$  as aqueous solutions or, in the case of pinene, as a pure standard. They were then nebulized and vaporized in a stream of zero air at  $1 \text{ L min}^{-1}$  and introduced upstream of the filter in the middle of the duct via a PTFE capillary heated to  $100^\circ\text{C}$ . The concentration of standard solutions was such as to obtain a mixing ratio of approx. 100 ppb in a  $400 \text{ m}^3 \text{ h}^{-1}$  flow. The volatiles were sampled simultaneously upstream and downstream of the filter from the middle of the duct at  $5 \text{ L min}^{-1}$  through PFA lines and subsampled from either one or the other into a proton-transfer-reaction quadrupole mass spectrometer (PTR-QMS 300, Ionicon Analytik, Austria) via a PFA line heated to  $100^\circ\text{C}$  (see Fig. 3). The instrument was operated at a reaction chamber pressure of 2.4 mbar,  $100^\circ\text{C}$ , 450 V, resulting in  $E/N$  of 104 Td. Besides the primary ion ( $\text{H}_3\text{O}^+$ ) and the water cluster ( $\text{H}_2\text{O}\cdot\text{H}_3\text{O}^+$ ), the analyser was set to monitor the ions at  $m/z$  corresponding to the protonated molecules, *i.e.* 45, 47, 60, 61, and 137. The raw data were processed using the PTR-MS Viewer 3.4 software.

The tested activated carbon filters were commercially available models intended for use in recirculating kitchen hoods. Two of the filters, shown in Fig. S6, were the ones used in the test kitchen experiments (henceforth designated as 'used'). Additionally, new units of the same type as the ones used in the test kitchen experiments were also tested.

Rather than measuring the breakthrough curves for the five VOCs standards [22] it was deemed that establishing the  $E_I$  parameter, *i.e.* the



**Fig. 3.** The filter capture efficiency test setup. It was comprised of a heated transfer line (1), a liquid calibration unit (2), a zero air generator (ZAG) (3), a liquid VOC standard (4), a variable-speed inline fan (5), a PFA sleeve (6), a filter housing with an activated carbon filter (7), a PTR-QMS instrument (8), and a flow meter (FM).



initial removal efficiency of the activated carbon filter measured at low (<1 ppm) challenge concentration [23] was more representative for the performance of the rangehood filters used in the test kitchen experiments. Each measurement began with  $10 \mu\text{L min}^{-1}$  of HPLC-grade water being introduced into the system via the LCU, with the airflow at  $400 \text{ m}^3 \text{ h}^{-1}$ . The flow upstream and downstream of the filter was then sampled alternately until the ratio of the signal for the used standard in both flows  $C_D/C_U = 1 \pm 0.02$  following ISO 10121 [23], thus establishing the baseline. Subsequently, the standard solution was substituted for the pure water and the upstream flow was measured until the analyser's real-time response reached a steady state, at which point the sampling point was switched to downstream of the filter. This was then repeated after decreasing the airflow (and thus the dilution of the standard) to 400, 350, 300, 250, 180, and  $108 \text{ m}^3 \text{ h}^{-1}$ . Each measurement was carried out in triplicate for each of the four filters, except for the trimethylamine standard for which a single set of measurements was performed for each filter since it took over an hour to reach a steady state.

After subtracting the baseline, the initial removal efficiency at each airflow rate was calculated from the ratio of the VOC standard's concentration at steady state upstream ( $C_U$ ) and downstream ( $C_D$ ) of the filter:

$$E_l = \left(1 - \frac{C_D}{C_U}\right) \times 100\%.$$

The pressure drop induced by the filters as a function of airflow was established by measuring the gauge pressure between the upstream and downstream sampling points using the Swema 3000md micro-manometer and juxtaposing the results with a parallel set of measurements in which the filters were removed from their housings.

### 3. Results and discussion

#### 3.1. VOCs emission profile

The emission from the cooking experiment with the base ventilation, together with the chemical compounds that were assigned to particular ions, is shown in Fig. 4. It is dominated by alcohols which constitute nearly 80 % of the TVOC, followed by carboxylic acids and aldehydes.

The VOCs emission profile measured during the cooking experiments is, on the whole, consistent with the results of recent studies in which PTR-ToF-MS was used to measure indoor air during cooking [19,24]. In particular, in the HOMEChem project [19] the model meal consisted of a rice and vegetable stir-fry, similar to the component of the meal used in our study, albeit with more cooking oil used per serving. The relative abundances of cooking-related VOCs from both studies are juxtaposed in Fig. 5. The salmon filets that were fried in our study can be identified as the source of trimethylamine with a high degree of confidence since it is a major volatile product of microbiological spoilage of chilled salmon [25]. Likewise, the salmon filet likely contributed to the emission of butyric acid and other carboxylic acids [26]. Another notable difference between the two VOCs emission profiles is the fact that we did not report the mixing ratio of acrolein which is released during frying with rapeseed oil [27]. While we did detect the  $\text{C}_3\text{H}_5\text{O}^+$  ion, we could not confidently assign it to acrolein due to the likely overlap with the fragment of propionic acid [28].

The high emission of ethanol is notable. Studies involving the analysis of volatiles from frying often list carbonyl compounds as the dominant VOC emission [18,29–31]. Others have indicated that the emission profile depends greatly on the type of cuisine, in many cases with the dominant emission of alkanes [32], possibly attributable to gas fuel rather than to foodstuffs. It should be noted though that studies relying on adsorbents for sampling of volatiles are impacted by low recovery rates in the case of ethanol [33,34]. However, in two recent studies involving PTR-ToF-MS measurements of cooking-related VOCs

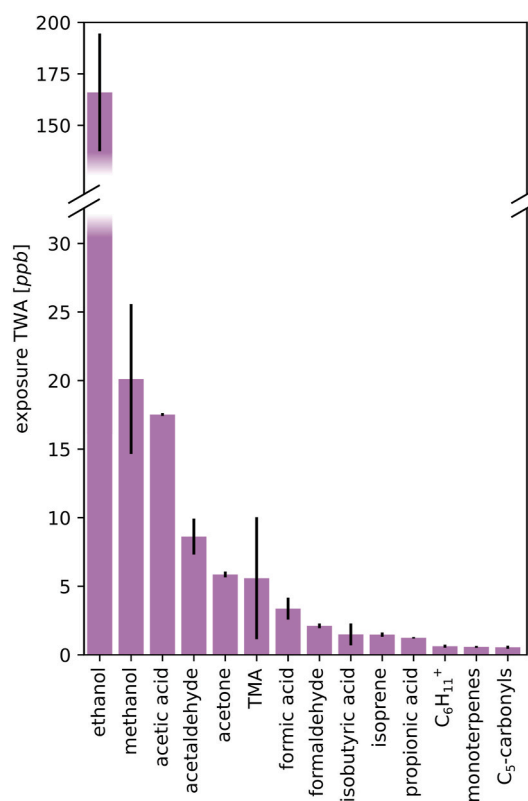
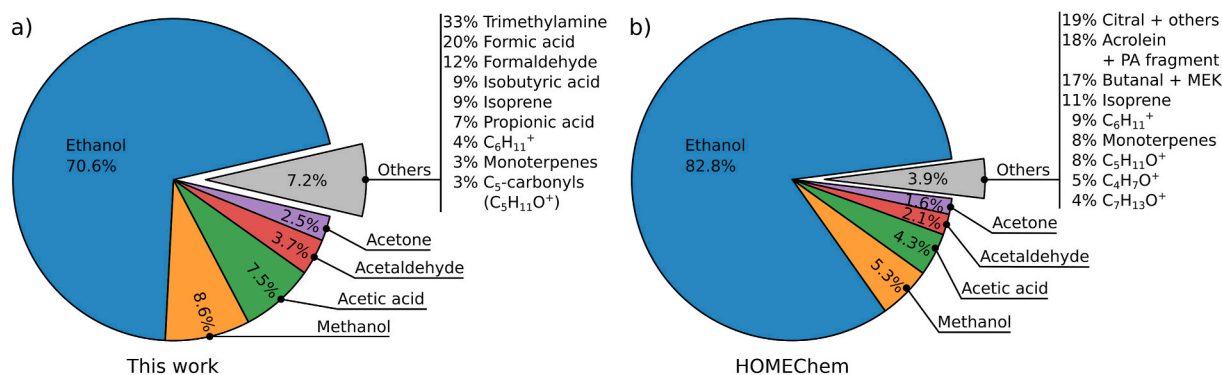


Fig. 4. Time-weighted average of the mixing ratio of monitored ions in the middle of the test kitchen (P2) during cooking with base room ventilation (scenario P2\_S0), and the compounds assigned to the ions. Error bars denote SD ( $n = 3$ ).

in the indoor environment ethanol was listed as the main compound, accounting for more than 80 % of the measured volatiles, both in the domestic and professional kitchen environment [19,24]. In both cases, the authors attributed some of the ethanol emission to cleaning products and sanitisers, noting however the major increase in ethanol mixing ratios during stir-fry cooking. In our study, the measurement periods did not encompass the cleaning that was carried out between the experiments eliminating that as a potential source, and yet ethanol accounted for over 70 % of the TVOC. While Arata et al. indicated that ethanol was an ingredient of a stir-fry sauce used during cooking [19], this, again, was not the case in our experiments. Still, alcohols constitute the majority of emissions from cooking vegetables [18], and there are further indications that the  $\text{C}_2\text{H}_7\text{O}^+$  ion, assigned here to ethanol, is the main product of the thermal oxidation of fatty acids [35]. Ethanol, alongside methanol, was also the most abundant VOC in the headspace of modified atmosphere-packaged salmon analysed using SIFT-MS [26]. Another recent study also showed that the emissions from cooking a chicken and vegetables stir-fry to be dominated by alcohols (70 % of VOCs emissions), albeit with methanol, rather than ethanol, reported as the most abundant compound [36].

Another takeaway from examining the baseline emission profile (*i.e.* without additional ventilation) is that the concentration of most of the monitored compounds did not exceed their respective irritating concentration thresholds [20,37]. Here the exception was acetic acid, the peak concentration of which exceeded the threshold of 25 ppb [20] during cooking in scenarios in which kitchen hoods were not used, although by a small margin. Likewise, of the monitored compounds, only the concentration of ethanol and TMA exceeded their respective odour thresholds (see Table S2 in the SI). The latter did so consistently in nearly all tested scenarios due to its particularly low threshold of  $0.8 \mu\text{g m}^{-3}$  [20].



**Fig. 5.** Comparison of the relative abundance of the 13 main compounds emitted during frying in this study (a) and in the HOMEChem study [19] (b). The percentages in the 'Others' categories are the percentages of the respective pie slices in subplots (a) and (b), not of the total.

### 3.2. Factors that affect the resident's exposure to VOCs during and after cooking

One should be careful not to draw conclusions based solely on the TVOC TWA exposure values shown in Table 2, mostly because of the high standard deviation on triplicate experiments for individual chemical compounds, as shown e.g. in Fig. S2 in SI. A majority of this variability seems to stem from the inherent inconsistency of the cooking ingredients. The airflow pattern in the room is also an important factor.

First, we consider whether the emission from the cooked food that was transferred to the table in the middle of the test kitchen was a major contribution to the overall TVOC emission. Based on the emission profiles of particular compounds this does not seem to be the case. While there are significant differences between the experiments in which the cooked dish was either sealed or left exposed, they are not consistent. In some cases, the overall emission during the experiments in which the cooked dish was sealed is higher than in experiments with corresponding setups and ventilation rates, but with the dish left exposed. This is e.g. the case for acetaldehyde, as shown in Fig. 6 (d), where no pattern emerged from comparing its emission profiles. A corresponding figure showing the emission profile of the main VOC, i.e. ethanol, is shown in Fig. S1 in the SI. They were higher when the food was sealed in some pairs of scenarios, while the inverse was true in others. This points to the foodstuff as the major source of variability of individual measurements.

**Table 2**

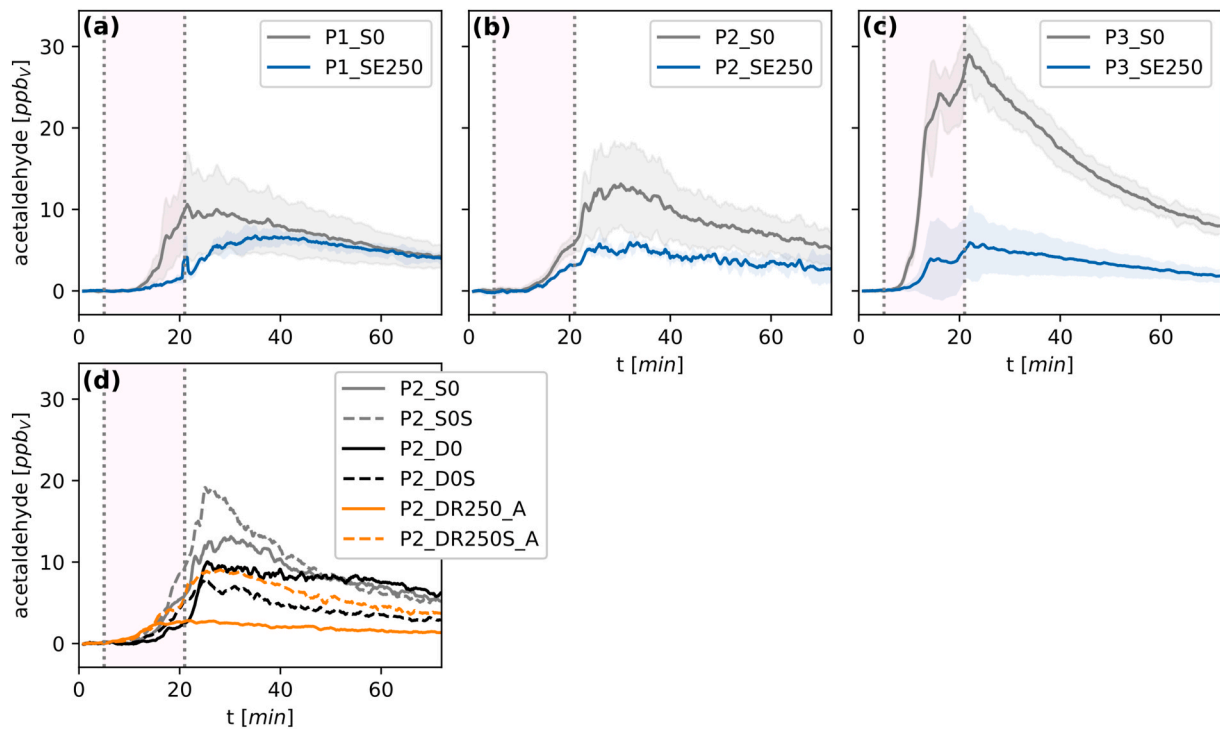
TVOC time-weighted (68 min) average exposure during the conducted experiments. Mean result  $\pm$  SD ( $n = 3$ ). Refer to Table 1 for further details on the designations.

Designation	Setup	Mode	TVOC TWA [ppbv]
P1_S0	Standard	n.a.	118 $\pm$ 27
P1_SE180	Standard	Extracting	64 $\pm$ 21
P1_SE250	Standard	Extracting	105 $\pm$ 12
P2_S0	Standard	n.a.	235 $\pm$ 42
P2_S0S	Standard	Extracting	229.8 $\pm$ 8.7
P2_SE180	Standard	Extracting	75 $\pm$ 33
P2_SE250	Standard	Extracting	97 $\pm$ 23
P2_SR180	Standard	Recirculating	188.6 $\pm$ 5.7
P2_SR250	Standard	Recirculating	135 $\pm$ 18
P2_SR180_A	Standard	Recirculating	242 $\pm$ 22
P2_SR250_A	Standard	Recirculating	121 $\pm$ 24
P2_D0	Downdraft	n.a.	197 $\pm$ 61
P2_D0S	Downdraft	n.a.	100 $\pm$ 22
P2_DR250	Downdraft	Recirculating	144 $\pm$ 24
P2_DR180_A	Downdraft	Recirculating	238 $\pm$ 25
P2_DR250_A	Downdraft	Recirculating	101 $\pm$ 82
P2_DR250S	Downdraft	Recirculating	131.6 $\pm$ 5.7
P2_DR250S_A	Downdraft	Recirculating	94 $\pm$ 22
P3_S0	Downdraft	n.a.	130.2 $\pm$ 3.2
P3_SE180	Downdraft	Extracting	119 $\pm$ 29
P3_SE250	Downdraft	Extracting	84 $\pm$ 30

Likewise, if the emission from the cooked meal was a major contribution to the overall exposure, we would expect the temporal emission profiles of particular compounds measured at measuring points P1 and P2 to differ significantly. There would likely be a marked increase in the measured signal as the exposed food is moved closer to the table and thus the inlet of the sampling line during the measurements at P2. This was not the case. At all three sampling points, we saw that the emission of acetaldehyde increased as the oil on the pan (its likely source) was heated and frying commenced. Similarly, in Fig. S1 we can see that the ethanol emission profiles at all three sampling points match, with a marked increase towards the end of the cooking, coinciding with the thawing of the frozen rice and vegetables stir-fry mix. The exception here is the ethanol emission profile in the recirculating setups, which we discuss in section 3.2.

It seems that the cooked dish is not a major source of VOCs compared to the cooking process, and so henceforth we will focus on scenarios in which the meal was transferred to the table in the middle of the room and left exposed since this is more realistic when considering the exposure of the room's occupant. The main source of variability of the measured TVOC is attributable to the foodstuffs used for cooking the model meal. Some of the measured VOCs, such as acetaldehyde or isobutyric acid, were characterized by a relatively low standard deviation in the triplicate measurements. In the case of the former, we assume that its main source was the rapeseed oil, measured in exact amounts in each experiment. In the case of isobutyric acid, the main source is likely the fat contained in the salmon filets. While the amount of fat in the particular portions differed, leading to noticeable differences between the replicates, on the whole, we would not expect the fat content to deviate too far from the approx. 16 % declared by the manufacturer. Unfortunately, this is not the case with the products of microbiological spoilage of fish. While in all experiments the salmon filets were before their *best before* date, how close they were to that date differed. The concentration of trimethylamine in raw salmon can increase by more than an order of magnitude within ten days of refrigerated storage in modified atmosphere packaging [38]. It is thus not surprising that its emission measured in the test kitchen experiments differed greatly between the experiments and between individual replicates (see for instance the SD for TMA in Fig. 5). Similarly, based on the ethanol and methanol emission profiles we can conclude that the frozen rice and vegetables mix was their major source. However, according to the observations of the researcher tasked with cooking the meal in all the experiments, the proportion of the ingredients in the pre-packaged mix, i.e. the ratio of rice to vegetables in the package, varied due to manufacture differences.

Likewise, by comparing subplots a : c in Fig. 6 we can see the notable effect of the airflow within the room on the measured exposure to VOCs. With no additional ventilation, the majority of cooking emissions were drawn towards the ceiling extract (Fig. 6c). Notably, in none of the experiments was the cook's overall exposure (P1) significantly higher than

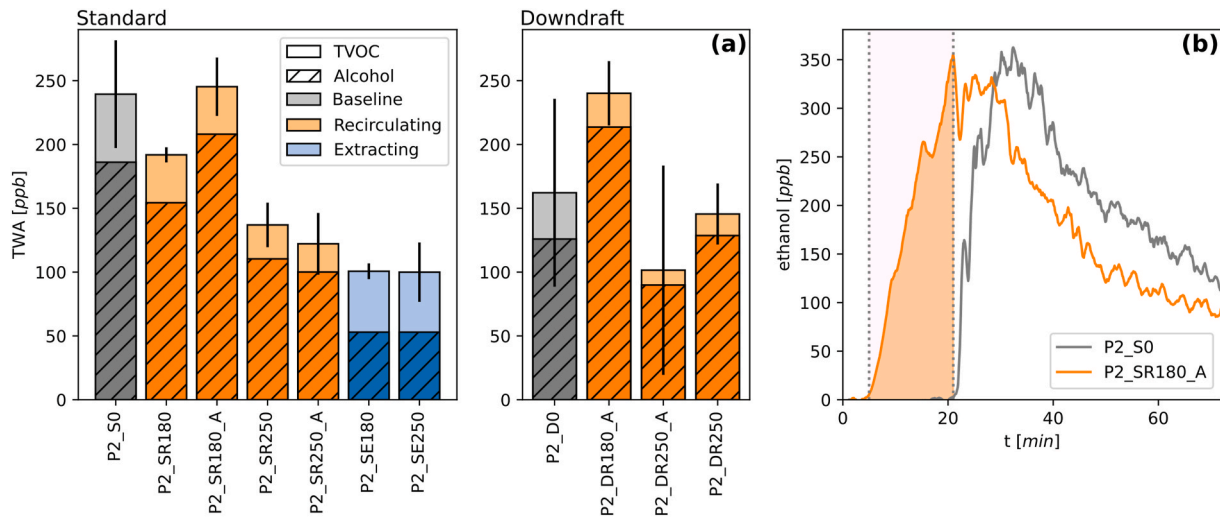


**Fig. 6.** Acetaldehyde emission profiles in different scenarios: measured at three different sampling points (P1, P2, P3, shown in subplots (a), (b), and (c), respectively), and at sampling point P2 (subplot (d)) during experiments in which the food was either sealed (dashed line) or left exposed (solid line). Solid and dashed lines denote mean measurement and the shaded areas, omitted in subplot (d) for clarity, show SD ( $n = 3$ ). The shaded area between the two dotted vertical lines indicates the cooking period.

the occupant’s exposure (P2). That was true even in scenarios in which the cooking hood was not used, with airflow likely affected by cooking-induced convection, the base ventilation through the ceiling exhaust, and even by the researcher’s movements during cooking [39]. As shown in Fig. 1, when either one of the setups was operated in the recirculating mode, the outlet of the recirculating flow was located away from the cook’s position. This induced the mixing of air in the test kitchen and hastened the moment at which the cooking emissions that were not captured by the filters reached sampling point P2 in the middle of the room.

### 3.3. Resident’s exposure to VOCs during and after cooking

The extracting setups outperformed the recirculating ventilation solutions in reducing the occupant’s overall exposure to cooking-related VOCs, as shown in Fig. 7a. Notably, alcohols constitute a higher proportion of the TVOCs in the case of recirculating setups, than in the case of the extracting setups. Further, in cases where each of the two setups was operated in the recirculating mode at  $180 \text{ m}^3 \text{ h}^{-1}$ , the exposure with kitchen hoods operated in the recirculating mode with afterrun exceeded the exposure with no additional ventilation.



**Fig. 7. (a):** Occupant’s exposure measured in the middle of the test kitchen (P2) in different ventilation scenarios; exposure to alcohols (sum of ethanol and methanol) is indicated by the darker, hatched area of the cumulative bars indicating exposure to TVOCs; error bars denote TVOC exposure SD ( $n = 3$ ). **(b):** Temporal profiles of the occupant’s exposure to ethanol measured in the middle of the test kitchen (P2) in two different scenarios; the shaded area highlights the difference between the two measurements during cooking.

In the previous section, we discussed how this might be due to the variability of the samples (foodstuff) or changing airflow patterns within the test kitchen. However, we did not observe such an effect in the same measurements for isobutyric acid, the emission of which was also sample-dependent in most cases (see Fig. S2 in SI). In fact, the exposure to isobutyric acid with kitchen hoods operated in the recirculating mode seems to be on par with the exposure with hoods operated in the extracting mode. This could suggest that the initial removal efficiency of the air filters used in the setups operated in the recirculating mode might affect the occupant's exposure to particular VOCs. If the filters are not particularly efficient in removing alcohols, then the increased air mixing induced by the recirculation might indeed reduce the time needed by that particular cooking emission to reach the middle of the room. This would, in turn, increase the average exposure time during the experiment.

However, in Fig. 7b we can see that when the cooking hood is operated in the recirculating mode, the ethanol mixing ratio measured at P2 increased before cooking commenced, that is immediately after the cooking hood was turned on. This could be an indication that induced airflow through the filters led to the desorption of the initially removed alcohol into the room, increasing the overall exposure measured from the moment the kitchen hood was turned on. Notably, the filters used in the test kitchen experiments were not purged between measurements in order to emulate real-life conditions as closely as possible.

We observed no major difference between the VOC capture efficiency of the 'standard' setup operated in the extracting mode at  $180 \text{ m}^3\text{h}^{-1}$  ( $67\% \pm 16\%$ ) and  $250 \text{ m}^3\text{h}^{-1}$  ( $57\% \pm 13\%$ ).

### 3.4. The initial removal efficiency of the activated carbon filters for kitchen hoods operated in the recirculating mode

The impact of the initial removal efficiency of the filters for different VOCs on the measured exposure within the test kitchen was difficult to assess in the presence of other potentially confounding variables discussed in section 3.2. This is why a separate experimental setup was used to test the performance of the filters in isolation. Five VOCs belonging to different chemical classes were selected from among the compounds assigned to the cooking emissions measured during the test kitchen experiments. It should be noted that the 'used' filters were in use in the test kitchen for a total of 5.2 h and 9 h for filter #1 and filter #2, respectively, which is only a fraction of their intended lifetime (the manufacturers recommend replacing them after 1 year of household use). Thus, it is likely that any observed differences in their performance are due to the variability of the filters themselves. The pressure drop induced by the individual filters at the measured airflow rates is shown in Fig. S4. The air passed through the filters was at  $44.9\% \pm 5.5\%$  p. RH and  $23.8\text{ }^\circ\text{C} \pm 1.4\text{ }^\circ\text{C}$ .

The initial removal efficiency of the filters as a function of airflow is shown in Fig. 8. All four tested kitchen hood filters had a removal

efficiency of over 80 % for acetic acid, which should be indicative of the removal efficiency for carboxylic acids in general. That is in line with the butyric acid removal results from the test kitchen experiments with the ventilation setups operated in the recirculating mode (see Fig. S2). Conversely, the acetaldehyde removal efficiency was  $<10\%$  in the best-case scenario at the lowest flow rate. The low adsorption capacity of activated carbon for acetaldehyde is a known issue [40,41], stemming primarily from its polarity [42,43]. This is unfortunate since exposure to acetaldehyde is known to have adverse health effects and the compound is considered a possible human carcinogen [44]. Its efficient removal by activated carbon filters requires specific pore sizes and the introduction of particular functional groups [45]. Monoterpenes are characterized by low polarity and are efficiently removed by the activated carbon filters, as was the case with pinene both in our experiments and in previous studies [46]. However, the removal efficiency for the polar trimethylamine was similar to that of pinene. This can be attributed to the use of activated carbon that was modified specifically for increased adsorption or the highly malodorous trimethylamine [37] and might be expected in filters intended for use in recirculating kitchen hood filters [47]. In general, the filters performed well in removing potential odorants such as TMA and pinene.

This, however, was not the case with ethanol: here, the initial removal efficiency ranged from 63 % to less than 10 % depending on the filter and airflow, as we expected based on the test kitchen experiments involving hoods operated in the recirculating mode. The poor performance of activated carbon filters in removing alcohols is also supported by the literature [46]. A further experiment in which the supply of the ethanol standard was stopped, but the airflow was kept on showed that the mixing ratio of ethanol measured downstream of the filter, shown in Fig. S3, took several hours to reach the background level, indicating substantial desorption from the filter. A similar effect was observed by Popescu et al. [48] who reported poor ethanol capture efficiency of activated carbon filters for ethanol and reported the ratio of ethanol concentration downstream of the filter to that upstream of the filter to be higher than 1 after measuring for a period of time. Wisthaler et al. [49] noted that a sorption-based gas filter reduced peak ethanol levels, but the subsequent re-emission decreased the overall removal efficiency. This supports the observation that the early increase of measured alcohol concentration in the test kitchen when the ventilation operated in the recirculating mode is turned on, as shown in Fig. 7b—is likely caused by desorption from the filter. Thus, it would seem that while the recirculating kitchen hoods equipped with activated carbon filters might to some extent mitigate the short-term exposure to spikes of alcohol emission during cooking, their efficiency in reducing the overall residents' exposure in small apartments is low.

Based on Fig. 8 it can be seen that the initial removal efficiency tends to decrease with the increase of airflow. While this produces a small effect in the case of acetic acid where the overall initial removal efficiency is approx. 90 %, it is noteworthy in the case of pinene (approx. 20

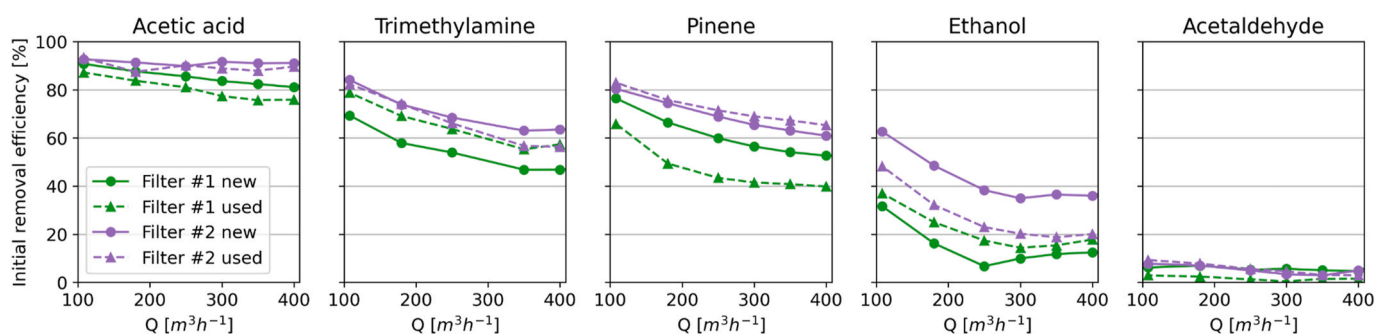


Fig. 8. The initial removal efficiency of activated carbon recirculating kitchen hood filters from two different manufacturers with five different chemical standards, as a function of airflow. One filter of each pair was new, and the other used for a relatively short period of time during actual cooking. Plotted points are the average of  $n = 3$  measurements with the exception of trimethylamine, in which case they are the result of a single measurement.



%–40 % difference between the  $E_f$  at 108 and  $400 \text{ m}^3\text{h}^{-1}$ , depending on the filter), TMA (approx. 30 % difference), and ethanol (approx. 43 %–61 % difference, depending on the filter). The initial removal efficiency experiments were performed at a single pass which might not be directly indicative of the performance of the recirculating ventilation at high airflow rates in small apartments. However, from the VOCs removal standpoint, there seems to be no clear benefit in increasing the recirculating kitchen hood's airflow beyond  $250 \text{ m}^3\text{h}^{-1}$ , provided that this is sufficient to capture the majority of cooking fumes.

### 3.5. Limitations

The aim of this study was to assess how different kitchen ventilation configurations affect the resident's exposure to cooking-related volatile organic compounds, particularly in small urban dwellings. To this end, the experiments were designed to emulate real-life conditions as closely as possible in terms of the experimental setup, *i.e.* the size and layout of the test kitchen, the cooking procedure representative of typical Norwegian cooking, and the use of residential ventilation solutions. This approach has some inherent limitations. As it became apparent during the measurement campaign, sample variability, *i.e.* the variability of the cooking ingredients, had a major impact on the measured emissions of VOCs, even with a highly standardized meal composition and cooking procedure. An ancillary test revealed, for instance, that sautéing a salmon fillet skin-side down, as opposed to a skinless salmon fillet that was being sautéed in this study, produced nearly an order of magnitude higher emissions.

Likewise, the frying temperature is in real life seldom controlled as diligently as during experimental work. Excess heat could lead to increased emission of semi- and low-volatile organic compounds, including carcinogenic polycyclic aromatic hydrocarbons (PAHs) [50], which were not measured in this study. This could in the future be addressed *e.g.* by using a PTR-ToF-MS equipped with the CHARON inlet [50] for the analysis of the organic fraction of the cooking aerosol. Additionally, the distribution of volatiles in the test kitchen could be more accurately mapped by distributing several photoionization detectors (PIDs) in a grid pattern and relating their TVOC measurements to the more detailed results obtained concurrently using a PTR-MS.

In the test kitchen experiments, the recirculating hood filters were mounted at a distance from the hoods and connected via a metal duct segment. While this configuration was supported by the manufacturers of both units and the filter housings for the ducts were bundled with the products, both hoods also featured a possibility to mount the filters directly after the fan. In such cases, the cooking fumes would pass through the filters at a higher temperature and lower relative humidity. In general, both lower RH and higher temperature would negatively affect the filters' performance [51,52].

Lastly, it should be noted that in order to test different scenarios in conditions resembling real-life cooking as closely as possible the number of replicates in the relatively time-consuming measurements was constrained to three. Thus, the obtained dataset is not ideally suited for multivariate statistical analysis focused on quantifying the effect of different factors. This could be addressed in the future *e.g.* by employing a dedicated laboratory setup similar to the one used in a recent study on the effectiveness of adsorption and photocatalytic filters in removing cooking emissions [53], allowing for a greater sample throughput, albeit arguably at the cost of representativeness for the home environment as a whole.

## 4. Conclusion

Based on experiments in which the meal was sealed directly after cooking, it can be stated that in the case of a typical Norwegian meal, cooked food on a plate by itself emits VOCs at a much lower magnitude than cooking the food on a stove. Both the inherent inconsistency of the cooking ingredients (*e.g.* fat content) and airflow conditions within the

test kitchen had a non-negligible impact on the measured exposure rates. Accounting for these, there does not seem to be a major difference in VOCs removal performance between kitchen hoods operated in the extracting mode at  $180 \text{ m}^3\text{h}^{-1}$  and  $250 \text{ m}^3\text{h}^{-1}$ . The cook's overall exposure was not higher than the exposure of a person sitting in the middle of the room.

However, the kitchen hoods operated in the recirculating mode did underperform compared to the ones operated in the extracting mode. This is in part due to the increased air mixing within the room induced by their operation but mostly results from the poor performance of the tested activated carbon filters in the removal of some of the cooking emissions. The configuration of the recirculating setup (either standard or downdraft) did not have a major effect on the occupant's exposure to cooking-related VOCs.

The activated carbon filters themselves, when tested using a dedicated experimental setup, performed relatively well in the removal of acetic acid and potential odorants, namely pinene and the particularly malodorous trimethylamine. In the case of ethanol, they exhibited low removal efficiency, and whatever fraction was initially removed was desorbed from the filters over an extended time. The initial removal efficiency for acetaldehyde, which constituted a non-negligible part of the measured cooking emissions, was particularly low. In terms of VOCs removal by charcoal filters, lower airflow rates are in general favourable, and the results indicate that there is no reason to use airflow rates exceeding the highest one tested, *i.e.*  $400 \text{ m}^3\text{h}^{-1}$ . The poor removal efficiency of alcohols, which constituted nearly 80 % of the measured cooking-related VOCs, explains the poor performance of the recirculating ventilation setups used in the test kitchen experiments. The desorption of alcohols from the filters further exacerbated this issue, as they were released into the room the moment the cooking hood was turned on.

Extracting ventilation solutions remain the most efficient way to remove cooking emissions by a large margin. Recirculating systems fitted with activated carbon filters are removing only some of the monitored cooking-related VOCs. This should be taken under careful consideration alongside data on particle removal in ongoing and future revisions of the relevant building codes.

### CRediT authorship contribution statement

**Wojciech Wojnowski:** Writing – original draft, Visualization, Methodology, Investigation, Formal analysis. **Aileen Yang:** Writing – review & editing, Methodology, Conceptualization. **Tomas Mikoviny:** Methodology, Data curation. **Armin Wisthaler:** Writing – review & editing, Supervision, Methodology. **Kari Thunshelle:** Writing – review & editing, Supervision, Project administration, Methodology, Funding acquisition, Conceptualization.

### Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

### Data availability

Data will be made available on request.

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not involved in study design, data collection, analysis, and interpretation, writing of the manuscript, nor in the decision to submit the article for publication.

## Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.buildenv.2024.111743>.

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