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Research Article

Assessment of Indoor Air Pollution From Cooking and Cleaning Activities in Italian Restaurant Kitchens

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This work assesses indoor air quality (IAQ) in 15 restaurant kitchens in Northern Italy by monitoring air pollutants from both cooking and cleaning activities. Volatile organic compounds (VOCs), aldehydes, ozone (O_3), nitrogen dioxide (NO_2), carbon dioxide (CO_2), and ultrafine particles (UFPs) were monitored across two seasons—winter and summer. Organic gasses were monitored using off-line techniques, whereas real-time monitoring was employed for inorganic gasses and UFPs. Contextual data were collected via checklists and video recording. Results showed higher O_3 levels in summer, whereas an opposite behavior was observed for UFPs, NO_2, and CO_2. D-limonene, propionaldehyde, and acrolein were the most abundant compounds in both seasons. Most pollutant levels were below guideline values, despite the occurrence of short-term pollution peaks, especially for O_3, TVOCs, and NO_2. Notably, benzaldehyde levels exceeded short-term guidelines in more than 1/4 of the restaurants, whereas acrolein surpassed the short-term exposure limit in up to 67% of cases. High levels of benzene were also recorded in about 1/4 of cases. Formaldehyde guidelines were also exceeded in some instances. This study highlights cooking as the main pollution source, with washing activities contributing to could cause IAQ guideline values exceedances. Adaptive ventilation systems and separating cooking and washing can represent effective mitigation measures.

Keywords: cooking emissions; dishwashing; indoor air pollutants; inorganic gasses; occupational exposure; VOCs and carbonyls

1. Introduction

Indoor pollutants may arise from various sources, including indoor sources, human activities and outdoor infiltration [1]. Among indoor activities, cooking is one of the most relevant contributors to pollutant emission [2]. Literature studies showed that cooking releases pollutants into indoor air, with numerous factors influencing indoor air quality (IAQ). These factors encompass cooking styles, the equipment used, the use of spices and oils, and cooking temperature [3]. Cooking emissions typically consist of various organic and inorganic pollutants, such as polycyclic aromatic hydrocarbons (PAHs), volatile organic compounds (VOCs), aldehydes, ultrafine particles (UFPs), and fine particulate matter ($PM_{2.5}$) [4]. It has been observed that cooking fumes and vapors contain high amounts of VOCs and aldehydes especially when frying oil and food due to a series of chemical reactions occurring at high temperatures [5]. Frying can also be responsible for large amounts of $PM_{2.5}$ emissions, contrary to other cooking methods like steaming and boiling [6]. Grilling is another important source of particle-phase pollutants, leading to high indoor concentrations of submicron particles and $PM_{2.5}$ [7]. Additionally, grilling can increase exposure to benzene and PAHs [8].

Commercial cooking can lead to higher indoor air pollution (e.g., PM_{10} and VOCs) than domestic cooking [9]. It can also be an important contributor to ambient VOC and $PM_{2.5}$ emissions in urban areas [10]. In addition, professional cooks, who spend more time close to pollutant sources characterized by high-emission rates, are exposed to elevated indoor pollutant levels with respect to home cooks (i.e., nonprofessionals) [11]. It has been shown that restaurant workers, such as cooks and the whole kitchen staff, face high levels of indoor air pollutants in cooking vapors and fumes [12, 13].

In addition to cooking, activities such as washing (both by hand and by means of a dishwasher) and surface cleaning and disinfection (i.e., cleaning worktops, floors, and windows) can emit VOCs affecting IAQ in restaurant kitchens [14, 15]. Detergents used to wash dishes and clean worktops, windows and floors release volatile compounds, such as alcohols, aldehydes and terpenes, in the indoor environment [16].

It should be recognised that dishwashing detergents usually have a lower VOCs content than other cleaning products. However, these compounds can be primarily emitted from the dishwasher water into indoor air when the dishwasher is open [17]. Common VOCs released during these activities included limonene, α-pinene and p-cymene, often used as fragrances in detergents [18, 19]. These terpenes are of interest because they can react with ozone (O_3) to form reaction products either as gas-phase contaminants, such as formaldehyde (HCHO) or as secondary organic aerosols (i.e., organic UFPs) [20, 21]. To the best of our knowledge, only a few studies in Europe have focused on indoor pollutant emissions and IAQ in restaurant kitchens. These studies have typically focused on personal exposure to specific pollutants such as PAHs, aldehydes and inorganic gasses such as nitrogen dioxide (NO_2) [22, 23] showing that frying is a major contributor to occupational exposure in European restaurant kitchens [23]. Studies in Norwegian restaurants revealed the release of naphthalene and aldehydes during meat frying. Furthermore, frying on a gas stove resulted in higher occupational exposures than frying on an electric stove [22].

To date, studies on dishwashing emissions were predominantly carried out in domestic kitchens [15]. The limited research conducted in professional kitchens has addressed the biological contamination and dish sanitation in relation to the type and characteristics of dishwashing machines [24, 25], but not IAQ. Considering the above, the novelty of this work lies in a broad-spectrum approach to assess IAQ in professional kitchens. In other words, these indoor environments were examined by expanding the range of the investigated contaminants, focusing not only on pollutants typically associated with cooking processes but also on those usually linked to washing and cleaning activities (e.g., terpenes). Hence, this approach could help in assessing the contribution to the increase in the formation of secondary reaction products, especially HCHO. The main goal of this study is therefore to fill the gaps of knowledge regarding the combined impact of cooking, washing, and cleaning activities on the IAQ in restaurant kitchens, to better understand which pollutants, activities, and exposure situations may pose health risks for kitchen workers. The seasonal differences between pollutant concentrations and several specific characteristics of the restaurants were also considered. In this way, it was possible to identify risk management measures, not only to be implemented in Italian restaurants but also in professional kitchens of other countries that adopt similar types of cuisine as well as washing and cleaning products and habits.

2. Materials and Methods

2.1. Study Area: Geographical Location of the Restaurants. A total of 15 restaurants located in Northern Italy, near the cities of Como, Milan, Monza, Saronno and Varese, were recruited to assess the IAQ inside their kitchens (Figure 1). The study area, located in the north-central part of the Lombardy Region, is categorized in the C category according to the Köppen classification climate system. More precisely, based on the location of the restaurants, it can be stated that the specific category is Cfa. This category refers to the humid subtropical climate, characterized by hot, humid summers and mild to cold winters, with precipitation occurring throughout the year. It should be emphasized that the proximity to the lake in the case of the city of Como and the urbanization in Milan could influence the local microclimates [26]. The average temperatures during the monitoring sessions ranged from 0°C to 6°C and from 20°C to 30°C in winter and summer, respectively. Average precipitations in the cold season ranged from 55 to 90 mm with occasional snow events, whereas in summer they ranged from 55 to 70 mm. Winds are usually very light, but stronger gusts can occasionally occur especially in the hot season. Relative humidity can be quite high in summer, often reaching 70%–80%, whereas winters are very damp, with 80%–90% humidity levels. Regarding air quality, in colder months Milan and surrounding areas are characterized by high air pollution levels due to heating, traffic, and industrial emissions (as an example, average PM_{10} levels ranged from 45–70 $\mu g/m^3$). Como and Varese tend to have better air quality (average PM_{10} concentrations between 30 and 50 $\mu g/m^3$), even though these two cities can experience elevated pollution levels during stagnant weather. In the hot season, the air quality is slightly better than in winter (for example, average PM_{10} concentrations ranged from 20–40 $\mu g/m^3$ in Milan and from 15-35 $\mu g/m^3$ in Como and Varese). However, it is important to note that, in the study area, hot seasons can be characterized by high levels of O_3.

2.2. Study Design. The IAQ survey was organised in two seasonal campaigns, one in winter (December 2021–March 2022) and one in summer (May 2022–July 2022). Each restaurant was investigated once per each season. The monitoring was performed during lunchtime, approximately between 10 AM and 3 PM, during both working days (the same eight restaurants in both seasons) and weekends (the

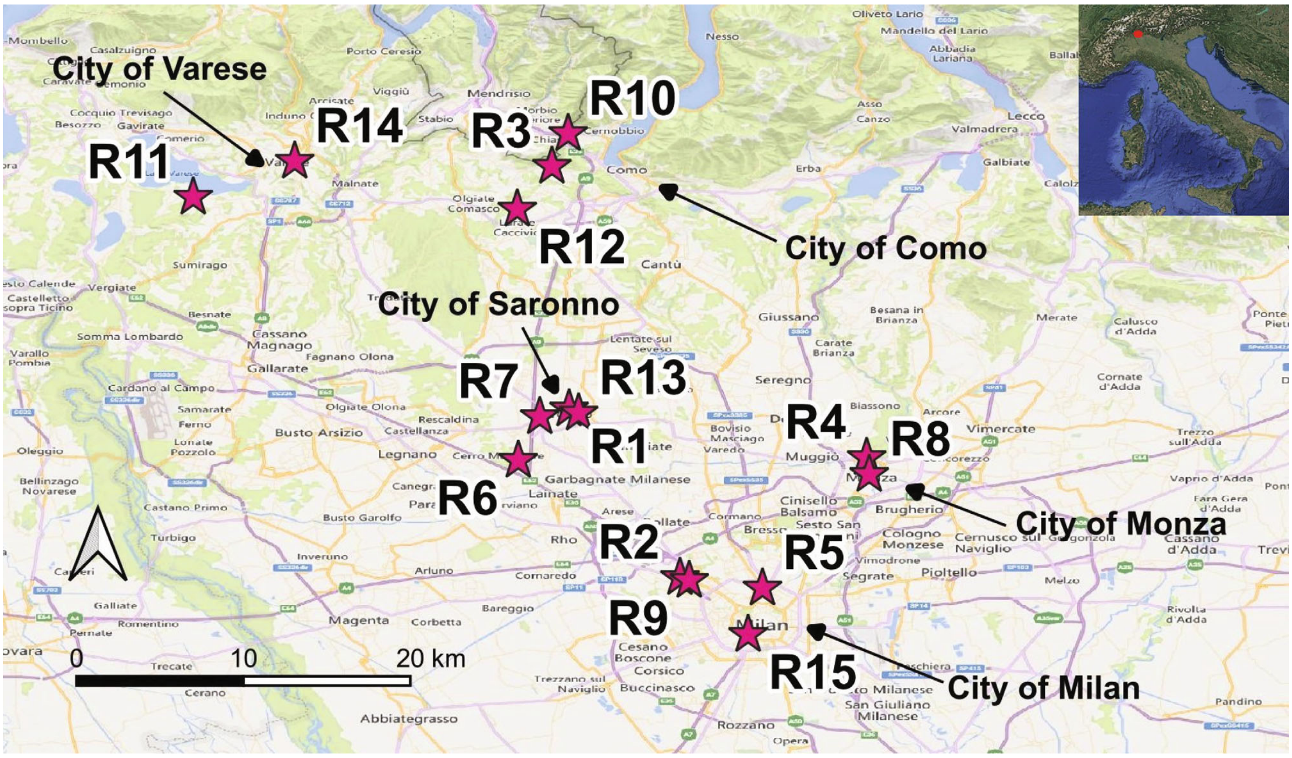

Figure 1: Geographical location of the restaurants (fuchsia stars). The study area, in Northern Italy, is indicated as a red point.

same seven restaurants in both seasons). Two types of restaurant layouts were observed, kitchens in which cooking and washing activities (i.e., both by hand and by dishwashing) took place in a single room (seven restaurants) and kitchens in which two separated rooms are specifically dedicated to these two activities (eight restaurants). Despite the different layouts, the sampling point was always chosen aiming to intercept the IAQ contribution of both activities (cooking and washing of dishes) in each kitchen. The cooking style was traditional Italian cuisine, and a few restaurants served pizzas that require cooking in a dedicated oven, but these were prepared and cooked in a separated location (outside the environment studied). A detailed checklist (Table S1) was filled in to obtain information about the general structural and operational characteristics of each restaurant, the presence of external sources and the specific characteristics of the kitchens. It is important to note that cooking conditions (e.g., types of hobs, type of oven), structural conditions (e.g., number of doors and windows, type of heating/cooling systems, type of extractor hood), and working conditions (e.g., number of workers and presence of dishwasher) remained the same between the two measurement campaigns. However, the nature of the study that was carried out in real scenarios meant it was impossible to control some influencing factors. Among these are the different types of cooking in the same restaurant (between the two measurement campaigns) because of seasonal variability of menus and the inter-restaurant variability due to the various culinary offerings [27]. The influence of time-activity patterns (including various kitchen activities such as cooking, washing, and cleaning) on IAQ was already evaluated in another recent study performed on the same restaurant sample, anyway [28]. As usual, since the study was carried out in real scenarios, several structural characteristics were different among the investigated restaurants. For instance, the minority (20%) of the restaurants had a kitchen volume less than 100 m^3, 40% between 100 and 200 m^3, and 40% greater than 200 m^3. Moreover, 33% of the kitchens had no windows or windows that could not be opened, 40% had only one window, 7% had two windows and 20% had three or more openable windows. All the kitchens had doors, but only 40% of them had doors that opened to the outside. There were extractor hoods in all the restaurants. As regard the possible infiltration of traffic-related air pollutants, nine restaurants (R1, R4, R5, R7, R8, R10, R11, R13, and R14) were located near roads; three restaurants (R2, R9, and R15) were close to both roads and industrial areas, whereas R3, R6, and R12 were located in rural areas.

2.3. Selected Pollutants and Sampling Strategies. Air concentrations of a variety of pollutants were measured indoors using a combination of on-line and off-line instruments placed on a tripod with sampling inlets at a height of 150 cm to represent the height of the workers' upper respiratory tract [29]. Specifically, the online monitoring covered the following pollutants: O_3, TVOCs, HCHO, UFPs, NO_2, and CO_2; whereas the off-line samplings were carried out on VOCs (i.e., benzene, toluene, (o-; m-; p-) xylenes, ethylbenzene, styrene, α-pinene, d-limonene, 2-butoxyethanol, and p-cymene) and aldehydes (i.e., HCHO, acetaldehyde, acrolein, propionaldehyde, benzaldehyde, glutaraldehyde).

These pollutants were chosen on the basis of previous work on indoor air pollutants released by cooking [11] and cleaning activities [30, 31] and their potential association with health effects [31]. Some inorganic and organic gasses, and UFPs as well, were selected accounting for previous studies investigating IAQ in public buildings [32] and private homes [33].

2.3.1. On-Line Monitoring. The time resolution of the on-line monitors was set to 1-minute. The average total sampling time was about 300 min, ranging from a minimum of 211 minimum to a maximum of 399. This timeframe was selected to encompass business hours (i.e., background concentrations and the different occupational activities performed in restaurant kitchens). O_3 was measured using the personal ozone monitor (POM, 2B Technologies), that has been certified as a Federal Equivalent Method by the U.S. Environmental Protection Agency [34]. TVOC concentrations were measured using an Aeroqual Series 500 monitor (Auckland, New Zealand), equipped with a photo ionization detector (PID) [35]. For HCHO monitoring, a real-time device (HAL-HFX205, Hal Technology, Cherry Avenue, United States) based on electrochemical sensing technology was used. In this regard, it is important to underline that this type of monitor is susceptible to numerous potential interferences, particularly from alcohols, as well-documented in the literature [36]. For this reason, real-time HCHO levels were corrected *a posteriori* based on off-line HCHO data collected by active sampling, as reported in the following paragraph. UFP concentrations were measured using the P-Trak 8525 (TSI Incorporated, Shoreview, Minnesota, United States), a portable condensation particle counter able to detect particle in the size range 0.02–1 μm [37]. NO_2 levels were recorded using the CairClip monitor (Cairpol; La Roche Blanche—France), a miniaturized device based on an electrochemical sensor consisting of three electrodes [38]. The CairClip is sensitive to some gaseous compounds, such as Cl_2, reduced sulfur compounds, and especially to high concentrations of O_3 [39]. CO_2 data were collected by means of a portable nondispersive infrared (NDIR) sensor (Telaire 7001) [40], connected with a datalogger (Hobo U12 Data Logger, Onset Computer Corporation, Bourne, Massachusetts, United States) capable of measuring also air temperature (T, °C) and relative humidity (RH, %). The instruments' location is reported in Figure 2. For further information see Table S2.

2.3.2. VOCs and Aldehydes Off-Line Sampling. The off-line samplings of VOCs and aldehydes were carried out focusing on the period of cooking and washing of dishes, for an average sampling time of 160 min to cover the period of service. VOCs were sampled using stainless steel tubes (PerkinElmer, Wilton, Connecticut, United States) packed with Tenax TA (200 mg, 60/80 mesh, Supelco Park, Bellefonte, Pennsylvania, United States) connected to a sampling pump unit set at a flow rate of 0.01 L/min for an average sampling time of 180 min [41]. Aldehydes were sampled using a sorbent tube packed with high-purity silica gel (ultralow background) coated with acidified DNPH (2,4-dinitrophenylhydrazine) (SKC, 150/300 mg sorbent, 20/40 mesh, 6 × 110 mm size) connected to a sampling pump set at a flow rate of 0.1 L/min for an average sampling time of 150 min [42] (Figure 2).

2.3.3. Additional Parameters. For each restaurant, the airflow of the exhaust hoods was measured using a Hygro-Thermo-Anemometer (HTA 4200, Pacer Instrument, Keene, New Hampshire, United States). The hood was divided into three sections, each of which was measured for 1 min and the data averaged to provide average velocity data. During the sampling, information on window opening, weather conditions, and daily cover counts were collected by the operator. In addition, two video cameras (model Dash Cam 45, Garmin Ltd., Olathe, Kansas, United States; model Action Cam HDR-AS50, Sony) were used to record real-time washing and cooking activities (e.g., type of cooking, turning on/off stoves, and dishwashers running and not).

2.3.4. Off-Line Samples Analysis

2.3.4.1. VOCs. After sampling, the substrates were stored in a refrigerator at 3°C until analysis, which was performed by an automated thermal desorber (ATD 400, PerkinElmer, Norwalk, Connecticut, United States) operating in the two-stage desorption mode and coupled to a gas chromatograph (GC, HRGC MEGA 2 series, MFC 800, Fisions Instruments) equipped with a flame ionization detector (FID, EL-980, Fisions Instruments) [43, 44]. The thermal desorption consisted of two steps: during the primary desorption, the sample is heated, and organic compounds are transferred from the tube to a cryofocusing trap by a flow of inert gas. The secondary desorption starts with a rapid heating of the cold trap, so that the cryofocused VOCs can be desorbed for a second time and injected onto the gas chromatographic column via a heated transfer line. For the separation of the analytes, an open tubular column was used, so that the carrier gas (He) flowed through the central space. Table S3 reports the operating parameters used for the ATD-GC-FID analysis. Moreover, detailed information about the whole analytical procedure (e.g., selectivity, linearity and sensitivity, and tube desorption evaluation) can be found elsewhere [15].

2.3.4.2. Aldehydes. Each sampled tube was stored in a cold and dark environment and then chemically desorbed with 5 mL of acetonitrile (ACN, HPLC PLUS Gradient grade, Carlo Erba) according to the reference method (ISO 16000-3) [45] and the mass of the desorbed DNPH derivative was noted. Subsequently, an aliquot of the extracted sample was transferred to a vial and analyzed by high performance liquid chromatography (HPLC, Nexera XR, LC-20AD XR, SHIMADZU) equipped with an autosampler unit (SIL-20A XR) with 20 μL loop and a photodiode array detector (PDA, SPD-M20A). The HPLC system was equipped with a reversed-phase octadecyl (C-18) column (ZORBAX SB-Phenyl 4.6 mm × 25 cm) and an ultraviolet visible (UV) absorption detector operating at 360 nm. In this way, the amount of ultraviolet or visible light that is absorbed by the components of a mixture is measured. The operating conditions of the HPLC system are shown in Table S4. An in-depth look at the method optimization and the

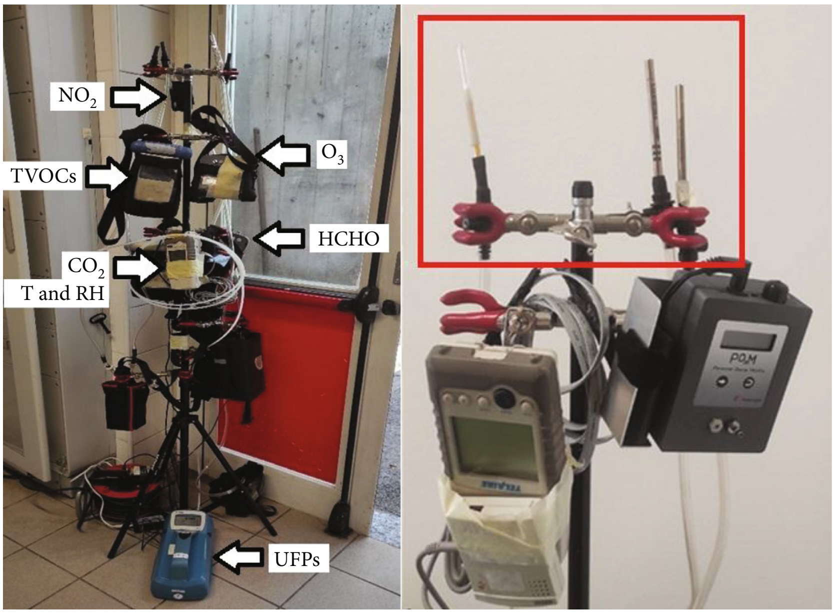

Figure 2: Instrumental setup used to measure air quality in the kitchens. The figure on the left shows the location of sampling devices in a restaurant. The right figure shows an enlargement (red rectangle) of the tubes used for off-line measurement of VOCs and aldehydes.

determination of organic molecules can be found in the supporting information (i.e., *Identification and quantification of organic molecules* [Table S5]).

2.4. Database Management and Statistical Analysis. After each sampling session, the video recordings were carefully examined to check and integrate the data collected by checklists with the aim to create a database for each restaurant and for each campaign. The timing of all real-time instruments and cameras was synchronized before each sampling session. In total, valid real-time monitoring data was collected for 4192 min in winter and 3301 min in summer. As regards off-line monitoring, one sample of VOCs collected in the winter campaign was lost during analysis ($N = 14$ and $N = 15$, in winter and summer, respectively).

The distribution of real-time and time-integrated data was verified by the Kolmogorov–Smirnov test. Based on the results of this test, seasonal differences were verified using the Mann–Whitney (M–W) test. Furthermore, the Spearman correlation coefficients were used to explore the correlations between pollutants and microclimatic parameters. In addition, to understand if the infiltration from the outdoor environment could affect IAQ, the concentration data of three typical outdoor pollutants (i.e., benzene, NO$_2$, and O$_3$) were retrieved from fixed monitoring stations managed by the regional agency for environmental protection (ARPA Lombardia) located as close as possible to the investigated restaurants. Based on the main cities areas, an overview of the distances between restaurants and the nearest fixed air quality monitoring stations is provided below: between 3 and 6 km for R2, R5, R9, and R15; between 2 and 9 km for R11 and R14; between 7 and 12 km for R3, R10, and R12; and between 1 and 6 km for R1, R6, R7, and R13. Then, the indoor/outdoor ratios (I/O) were calculated for each restaurant aiming to evaluate the impact of internal or external sources on indoor concentrations. To do this, outdoor concentrations were obtained by averaging hourly concentrations, approximating the start and end times with the closest time of the indoor measurement sampling time.

To assess IAQ, average data were compared with selected reference values. The IAQ guide values (GVs) or reference values are standards or benchmarks used to assess the concentration of specific air pollutants. They are typically set based on scientific research and health guidelines to indicate the level of pollutants that is considered safe or acceptable for human health. Specifically, for limit values equal to or less than 1 h, the average concentrations corresponding to the reference time of the guide value were used for comparison (e.g., for real-time measurements of NO$_2$ and HCHO). In the case of GVs referring to an 8-h period, an average of all the data collected during an entire 5-h sampling session were considered. The reference period of the limit values for benzene (one calendar year) did not allowed for a reliable comparison due to the fact that only two seasonal short-time measurements were available. To enhance the representation of the data and better manage large outliers, the "ggbreak" package was employed following the methodology outlined by Xu et al. [46]. The collected data were then analyzed using R Version 4.2.1. (R Foundation for Statistical Computing, Vienna, Austria) and SPSS Version 9.4 (SAS Institute, North Carolina, Cary, United States).

3. Results and Discussions

3.1. Restaurant Survey. Monitoring campaigns were carried out during the entire work session day, which can be conceptually divided into three phases: preservice, service, and postservice. During monitoring, the kitchen staff was always present in the investigated rooms, with an average of five people (ranging from 2 to 11). The average kitchen volume was 123 m^3 (ranging from 40 to 270 m^3). Most of the restaurants were located near traffic areas (80%), whereas 20% of them were in agricultural settings. In 13 restaurants out of 15, the kitchens had openable windows, whereas in the other two the windows were not openable. Most of the selected restaurants (14) were equipped with gas stoves and only one with an electric hob. The average number of customers served during the observation days was similar in winter (average 52; range 20 to 87) and in summer (average 50; range 13 to 120). A summary of the timing of the different activities performed in the investigated kitchens is reported in Table 1.

TABLE 1: Average time durations of activities carried out during monitoring in the 15 restaurants.

	Winter campaign average (range)	Summer campaign average (range)
Preservice (min)	99 (57–163)	108 (61–185)
Service (min)	109 (67–186)	89 (24–137)
Postservice (min)	68 (22–154)	92 (45–175)
Windows opening (min)	35 (14–262)	127 (98–431)
Boiling (min)	195 (58–392)	165 (27–301)
Grilling (min)	39 (15–199)	39 (5–152)
Frying (min)	24 (17–73)	13 (4–50)
Cooking activity (min)	231 (149–272)	173 (67–301)
Dishwashing activity (min)	179 (91–300)	147 (55–227)

Abbreviation: min = minutes.

3.2. IAQ. A summary of the indoor pollutant concentrations, microclimatic parameters (T and RH%) and hood extraction speeds measured during the two campaigns are shown in Table 2. First, most of the pollutants showed detectable concentrations, except for glutaraldehyde and styrene, which had approximately 50% below the limit of detection (< LOD). The very low concentrations of glutaraldehyde are in line with the findings of previous studies [47], whereas styrene was usually detected at higher levels than those found in the present work [11].

As expected, average O_3 concentrations were statistically higher in summer than in winter (0.019 ppm and 0.009 ppm, respectively), due to its photochemical origin [48]. The indoor air contamination by O_3 is in line with literature data showing typical concentrations from less than 0.005 ppm to over 0.05 ppm [49]. Likewise, the average values of TVOCs measured in summer were slightly higher than in winter (0.707 vs. 0.577 ppm, respectively). Observing also the standard deviations, medians and maximums, this fact could be attributed to peak episodes that could be linked to evaporative processes and an increase in oxygenated VOCs compared with winter [50]. As an example, TVOCs were positively correlated with 2-butoxyethanol (+0.62) in the summer campaign (Figure S3). In contrast, UFP levels were lower in summer than in winter. UFPs are typically produced by combustion sources (e.g., road traffic, power plants, residential heating, and cooking) and, to a much lesser extent, by ozonolysis reactions between O_3 and VOCs [51–53]. This seasonal behavior can be interpreted in the light of the higher air changes per hour in summer (due for instance to more time with windows open) than in winter (Table 1) [54]. The Spearman correlation revealed that UFPs were correlated with pollutants commonly linked with typical combustion sources, such as NO_2 in winter (+0.69), benzene and toluene in summer (+0.48 and +0.58, respectively) (Figures S2 and 3). The NO_2 concentrations were similar between the two seasons, although the levels measured in winter were slightly higher than in summer probably because of the lower air changes per hour and more time spent cooking in the cold season (Table 1). This is confirmed by CO_2 levels, which can be used as a proxy for ventilation and showed a similar seasonal trend, with an average level of 678 ppm in winter and 607 ppm in summer. The Spearman correlation results suggested that CO_2 was positively correlated with pollutants probably originated indoors, for instance d-limonene both in winter (+0.63) and in summer (+0.62), whereas it was negatively and highly correlated (−0.74) with a typical outdoor contaminant that is O_3 (Figures S2 and 3). Statistically significant differences ($p < 0.01$ and $p < 0.001$) between seasons were observed for the concentrations of all the above pollutants (Figure S1).

Among the selected VOCs, the highest average indoor concentration was measured for d-limonene, a typical monoterpene used as a fragrance in cleaning products, in both campaigns (on average, 31.4 and 15.3 $\mu g/m^3$ in winter and summer, respectively) [55]. This finding was supported by checklist data, in which d-limonene was confirmed to be a major component of dish detergents in most cases (83%). Higher average concentrations of the selected VOCs were always observed in winter than in summer, with statistical significance ($p < 0.05$) only for d-limonene, α-pinene, and BTEX (benzene, toluene, ethylbenzene, and xylenes) due also to the low sample size. *α-Pinene* is another natural active terpene widely used in cleaning products [14], whereas BTEX are more commonly associated with combustion sources such as vehicle traffic [56]. However, BTEX can also be emitted by cooking activities, especially grilling [57] but also frying because of the oil oxidation leads to the formation of alkadienyls, known precursors of these monoaromatic hydrocarbons [58]. The Spearman correlation results confirmed that several positive and significant associations were found between BTEX, as an example xylenes were highly correlated with benzene (+0.57) and ethylbenzene (+0.66) in the cold season and with toluene (+0.70) and ethylbenzene (+0.60) in the summer (Figures S2 and 3). In that season, elevated temperatures result in heightened UV radiation, leading to an increased photochemical production of O_3 outdoors. This, in turn, prompts the reaction of VOCs (especially terpenes) with O_3 causing a reduction of their concentrations indoors [59]. Another factor contributing to lower VOCs concentrations in summer is the increased natural ventilation, as more time is spent with windows open.

TABLE 2: Indoor concentrations of air pollutants, physical parameters (temperature and relative humidity), and hood extraction speed monitored in restaurant kitchens during the winter and summer campaigns.

	Winter						Summer					
Pollutants	**N**	**Min**	**Mean**	**SD**	**Median**	**Max**	**N**	**Min**	**Mean**	**SD**	**Median**	**Max**
O_3 (ppm) *	4189	< LOD	0.009	0.009	0.006	0.185	4192	< LOD	0.019	0.011	0.018	0.067
TVOCs (ppm) *	3471	< LOD	0.577	0.762	0.280	6.95	4174	< LOD	0.707	1.26	0.330	26.5
UFPs * 10^3(pt/cm^3) *	4105	1.42	62.5	69.7	32.5	497	4020	1.02	36.2	56.8	11.7	417
NO_2 (ppm) *	3942	< LOD	0.036	0.052	0.014	0.250	4327	< LOD	0.034	0.049	0.010	0.250
CO_2 (ppm) *	4189	387	678	220	623	1902	3972	419	607	132	574	1276
Benzene (μg/m^3)	14	< LOD	6.14	4.92	4.09	13.9	15	< LOD	0.895	1.01	0.589	3.53
Toluene (μg/m^3)	14	< LOD	18.5	16.6	9.31	53.2	15	< LOD	5.34	6.51	1.68	18.1
Ethylbenzene (μg/m^3)	14	< LOD	3.58	3.84	1.98	13.4	15	0.130	1.89	2.28	1.38	9.42
p-Xylene (μg/m^3)	14	< LOD	2.10	2.47	1.07	8.21	15	< LOD	1.43	3.25	0.123	9.94
m-Xylene (μg/m^3)	14	< LOD	6.04	6.12	4.22	23.4	15	< LOD	2.96	2.23	2.41	9.14
o-Xylene (μg/m^3)	14	< LOD	2.75	2.86	1.33	9.01	15	< LOD	2.00	1.64	1.91	6.71
Styrene (μg/m^3)	14	< LOD	1.09	1.18	< LOD	2.93	15	< LOD	0.766	0.964	< LOD	2.48
α-Pinene (μg/m^3)	14	< LOD	3.48	5.29	2.16	18.1	15	< LOD	0.897	1.11	< LOD	2.84
d-Limonene (μg/m^3)	14	< LOD	31.4	20.4	28.9	80.1	15	0.902	15.3	20.0	6.83	66.2
p-Cymene (μg/m^3)	14	< LOD	3.99	3.30	3.74	10.4	15	< LOD	2.20	2.80	0.932	7.98
2-Butoxyethanol (μg/m^3)	14	< LOD	21.7	34.7	6.80	114	15	0.156	10.2	23.2	2.08	91.2
Formaldehyde (μg/m^3) **	15	0.364	6.59	5.80	4.35	20.4	15	< LOD	4.71	3.79	3.85	15.2
Acetaldehyde (μg/m^3)	15	0.907	21.3	24.1	11.8	79.2	15	< LOD	15.2	12.6	9.87	45.4
Acrolein (μg/m^3)	15	2.81	25.7	25.0	16.2	82.2	15	10.4	34.5	25.7	23.0	99.0
Propionaldehyde (μg/m^3)	15	< LOD	25.7	26.6	16.1	78.2	15	< LOD	37.4	33.1	31.4	116
Benzaldehyde (μg/m^3)	15	< LOD	16.7	13.2	11.8	47.6	15	3.04	16.2	14.5	8.79	49.5
Glutaraldehyde (μg/m^3)	15	< LOD	2.01	1.13	< LOD	3.08	15	< LOD	3.57	1.48	3.28	5.54
Temperature (C°)	4191	9.83	20.2	3.19	20.7	25.5	4327	14.4	30.7	2.44	30.2	36.6
RH (%)	4191	21.7	40.1	9.80	40.4	67.5	4327	29.5	44.7	5.76	44.7	65.3
HES (m/s)	14	0.100	0.597	0.366	0.530	1.51	12	0.080	0.537	0.249	0.430	0.961

Note: The sampling times for VOCs and aldehydes are reported in Tables S6 and S7 in the Supporting Information. The sampling times for on-line monitoring are included in Tables S9 and S10 in the Supporting Information.

Abbreviations: HES, hood extraction speed; LOD, limit of detection; max, maximum concentration; min, minimum concentration; N, number of kitchens sampled RH, relative humidity; SD, standard deviation.

* First pollutants refer to parameters measured with direct-reading instruments.

**Data here presented for formaldehyde concerns only those time-integrated. More information about real-time data is available in the Supplementary Materials (Tables S9 and S10).

This augmented ventilation helps to dilute indoor air pollutants, especially when outdoor contamination is minimal [60]. Conversely, during the cold season, terpenes play a lesser role in reactions initiated by O_3 leading to higher concentrations of d-limonene and α-pinene indoors [19].

Regarding aldehydes, acrolein, and propionaldehyde emerge as the more concentrated compounds in indoor air throughout both seasons, reaching their highest values in summer (Table 2). This seasonal trend is also observed for glutaraldehyde, whereas HCHO, acetaldehyde, and benzaldehyde mean concentrations were higher in winter. Cooking, especially frying, stands out as a common source of carbonyls in kitchens, with greater prevalence in winter than in summer (see Table 1). Additionally, washing and cleaning activities can release terpenes into indoor air, acting as precursors to certain carbonyls when O_3 is available to initiate ozonolysis reactions [16, 58]. Aldehydes were positively correlated with RH, particularly propionaldehyde in winter (+0.58) and acetaldehyde and benzaldehyde in summer (+0.67 and +0.60, respectively). Humidity was closely related to cooking processes in kitchens, such as boiling, frying, and steaming, which generated high temperatures and promoted the formation of aldehydes as by-products [61]. As expected, HCHO, acetaldehyde, and acrolein were highly correlated with each other in winter (ranged from +0.56 to 0.79) probably due to their common origin (i.e., cooking), whereas only HCHO and acetaldehyde were correlated (+0.57) in summer possibly as to O_3-initiated reactions products (Figures S2 and 3).

Finally, Table 2 reveals a high variability (SD) for some aldehydes (i.e., acetaldehyde, acrolein, propionaldehyde, and benzaldehyde) and certain VOCs (e.g., toluene, d-limonene, and 2-butoxyethanol). This variability aligns with the differences between the investigated restaurants, showcasing

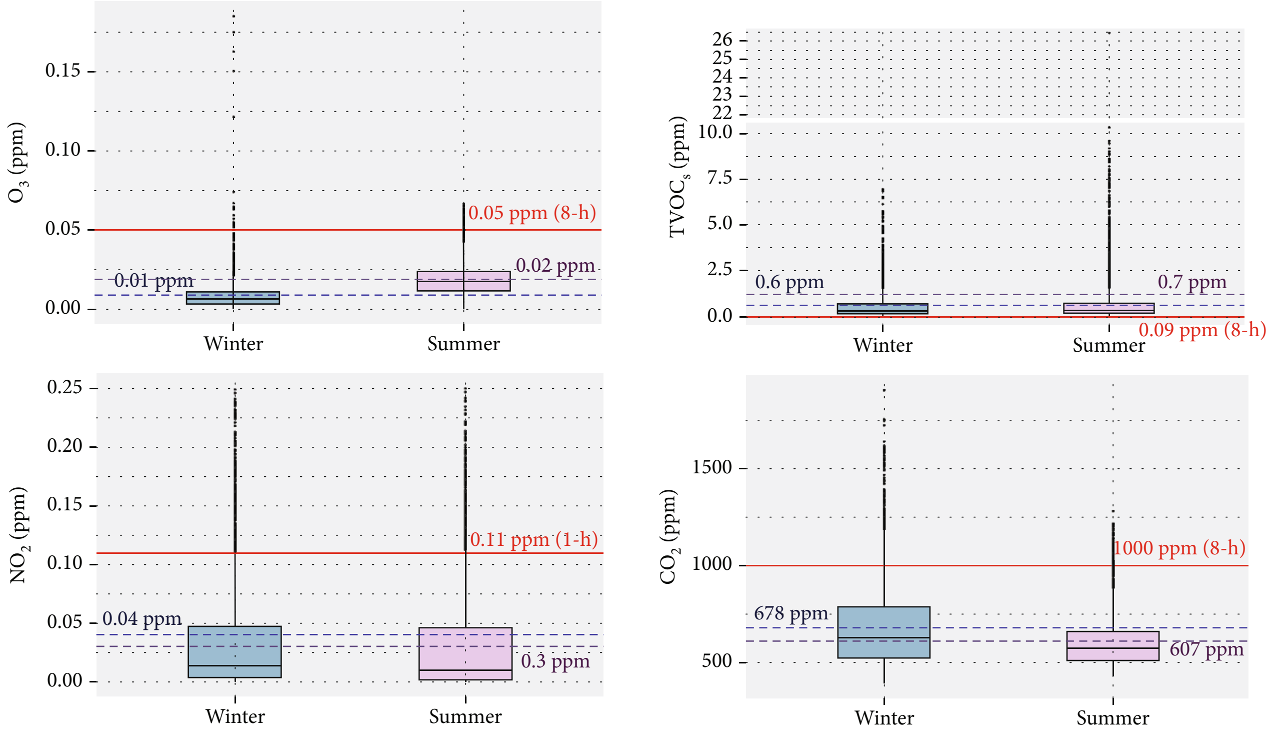

Figure 3: Boxplot of the winter (light blue) and summer (pink) measurements of the real-time pollutants and the corresponding IAQGVs (red line). The average of the winter (blue line) and the summer (purple line) campaigns is shown.

distinct concentrations between them (Tables S6 and S7). Several factors could contribute to these variations. Firstly, the positioning of the sampling point (as detailed in Paragraph 2.1.) might have led to higher concentrations of specific pollutants compared with others. For instance, the vicinity of the washing area exhibited higher concentrations of terpenes, such as d-limonene (+59% in winter and +24% in summer), in contrast to pollutants released during cooking activities, like toluene (+16% and +6% in winter and in summer, respectively) and acrolein (+32% and +33% in winter and summer, respectively) [17, 62]. Moreover, the size of the kitchen and the type of ventilation (natural or mechanical) may account for differences in compound concentrations [63]. In general, natural ventilation alone proves insufficient for controlling indoor air pollutants, necessitating the use of extractor hoods [64]. However, no statistical association was found between the data on hood extraction speed and the measured pollutant levels. Furthermore, distinct cooking methods can influence pollutant concentrations, with activities such as frying and grilling associated to elevated levels of VOCs and aldehydes [65, 66]. Regarding the influence of the outdoor environment on IAQ, the I/O ratios for O_3 (Table S11) were lower than 1 for all the investigated restaurants in both seasons, revealing that the infiltration played a key role both in winter and in summer. The I/O ratios for benzene and NO_2 (Table S11) were higher than one in 11 and seven restaurants during winter season while during summer in seven and 11 restaurants for benzene and NO_2, respectively. This result revealed that the external sources could have an impact on IAQ strictly related to the outdoor concentrations and air changes, although cooking activities would appear to be the main source for a major part of the restaurants.

3.3. IAQ Assessment. The IAQ of restaurant kitchens was assessed by comparing the measured pollutant concentrations with specific Indoor Air Quality Guideline Values (IAQGVs) or selected thresholds for irritation and respiratory effects (Table S8). These GVs, when not exceeded within the specified time interval, are expected to provide protection against possible health effects, ranging from the so-called critical effect to all conceivable adverse effects expected to occur at higher doses. It should be noted that for some compounds, the IAQ assessment was also not carried out due to a lack of IAQGVs (e.g., for UFPs).

3.3.1. On-Line Data. Figure 3 illustrates the distributions of all real-time data collected in winter (depicted in light blue) and in summer (depicted in pink), alongside the corresponding IAQGVs represented by the red line. An 8-h value was selected for O_3, TVOCs, and CO_2, whereas a 1-h value was selected for NO_2. For all pollutants, the maximum concentrations surpass the IAQGVs, indicating a possible exceedance of reference values. However, it is noteworthy that, among these pollutants, only TVOCs exhibit average concentrations higher than the selected IAQGVs. This can be attributed to cooking and washing activities, which release high amounts of various VOCs into the ambient air [67, 68]. Furthermore, the use of a short-term reference value for NO_2 allowed for an examination of concentration trends compared with the reference value (0.11 ppm) for each hour. It was observed that 33% of restaurants exceeded the GV in summer, whereas only 13% did so in winter (Tables S9 and S10). This exceedance is primarily associated with preservice and service activities characterized by intense cooking [69]. For HCHO, classified as a Group 1 carcinogen by the *International Agency for Research on Cancer* (IARC) [70], the WHO IAQGV of 0.08 ppm (100 μg/m^3) based on a reference time of 30 min [71] was selected as a benchmark. This value is considered to be protective against both chronic and acute health effects, starting from sensory irritation [72]. Three instances of exceedance of this threshold were observed among 30-min time-averaged concentrations, all within the same restaurant, along with 12 transient short-term peaks above the threshold in five restaurants as a whole. Lastly, some instantaneous exceedances of the CO_2 reference value of 1000 ppm, which is not health-based but a proxy for the minimum outdoor air ventilation rate per occupant required to control human-generated bioeffluents, occurred in seven restaurants in winter and in three restaurants in summer (Tables S9 and S10).

3.3.2. Off-Line Data—VOCs. Among the sampled VOCs, benzene is of particular interest because of its well-known carcinogenicity. The reference value, set at μg/m^3 as the annual limit value by the European Union, refers to a whole calendar year, whereas the available results are based only on a few hours of sampling [73]. Thus, no sound conclusions can be drawn, being the average of the available summer and winter data an improper surrogate of annual concentrations. However, our findings indicate that high levels of benzene (mean value of 6 and maximum of 14 μg/m^3—Table 2) were measured in winter. For several of the compounds investigated, the mean and maximum concentrations fall below the corresponding IAQGVs. Notably, d-limonene and α-pinene showed concentrations well below the critical exposure limits (CELs) derived within the EU-EPHECT (emissions, exposure patterns and health effects of consumer products in the EU) project, which were set at 90 and 45 mg/m^3, respectively, [74, 75]. Similar findings apply for p-cymene and 2-butoxyethanol, with DNEL values (derived no effect levels) for the general population set at 0.22 and 56 mg/m^3, respectively, [76, 77]. It is worth noting that terpenes can be involved in ozonolysis reactions leading to the production of gas- or particle-phase (e.g., UFPs) pollutants including HCHO and other VOCs recognized as respiratory irritants [77]. Furthermore, the concentrations measured for other VOCs were much lower than the corresponding IAQGVs. This is evident for *(o-; m-; p-)* xylenes, ethylbenzene, and styrene, all below the IAQGVs, consistent with findings from other studies, except for toluene, for which concentrations above 10 μg/m^3 were observed [78].

3.3.3. Off-Line Data—Aldehydes. For several of the aldehydes examined, the average and maximum concentrations are largely within reference values. In particular, the measured concentrations of acetaldehyde, classified by IARC as Group 2B, are well below the selected reference value (1.42 mg/m^3) [79]. Even for glutaraldehyde and propionaldehyde, the selected IAQGVs are widely respected (0.2 and 6.1 mg/m^3, respectively). Figure 4 shows the concentrations of benzaldehyde and acrolein compared with short-term reference values. Benzaldehyde was monitored at levels exceeding the Germain AIR (German Committee on Indoor Air Guide Values) GV-I reference value (Guide value I) but not the GV-II (Guide value II). In more detail, the GV-I is a precautionary guide value that describes the concentration of a substance in indoor air, for which or below which adverse health effects are not expected even after a lifetime of exposure, whereas the GV-II is an effect-related value requiring immediate action based on current toxicological and epidemiological knowledge. For the sake of completeness, GV-I was exceeded in 27% of restaurants in winter and in 33% in summer. These restaurants were characterized by intense grilling activity of the food, which could explain higher benzaldehyde concentrations than other restaurants [80]. The critical effects associated with GV-I refer to irritation of the eyes and mucous membranes of the respiratory tract [81]. However, no immediate resolution action is required, as the concentrations remain well below the GV-II level [82]. Anyway, as previously specified in the section dedicated to VOCs, for certain aldehydes it was chosen to compare the average of the summer and winter data with the annual reference value, as a surrogate and rough approximation of a possible annual average.

One significant finding of the present study is the frequent exceedance of the IAQGV for acrolein (21 μg/m^3 for short-term effects referring to a reference time of 1 h and derived in the framework of the EU-EPHECT project). Specifically, 40% of the restaurants exceeded the 1-h CEL in winter and a substantial 53% did so in summer (refer to Table S7). Moreover, a larger fraction of restaurants (73% in winter and 100% in summer) would be not compliant with the 8-h CEL, but this exercise should be considered precautionary, given the large difference between the actual sampling duration (2-h) and the 8-h reference period. Acrolein is an aldehyde formed by incomplete combustion of organic material and it is commonly released during frying activities [83]. High acrolein concentrations were measured in kitchens with intense boiling and frying activities, this last, especially in summer. It should be noted that the acrolein concentrations measured in the present study are above the IAQGV and relatively high compared with studies in restaurant kitchens in other countries [84, 85], but similar to those measured in domestic kitchens (24 μg/m^3) [86]. Exposure to acrolein at these levels might lead to adverse health effects, including irritation to the eyes and respiratory tract [87].

3.4. Strengths and Weaknesses of the Study. One of the strengths of this work is its comprehensive coverage of a large group of pollutants. In addition to investigating the levels released by cooking activities, the study addresses a literature gap by examining the levels of pollutants emitted also during the cleaning and washing activities performed in restaurant kitchens. Moreover, another advantage of this study is that the monitoring was carried out in real scenarios, allowing us to consider a broad spectrum of cooking

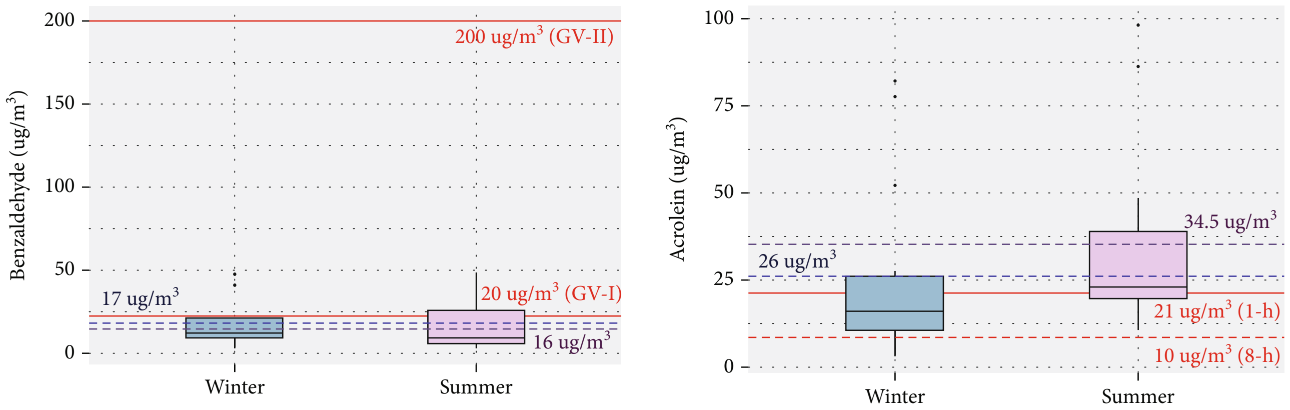

FIGURE 4: Boxplot of the winter (light blue) and summer (pink) measurements of the aldehydes and the corresponding IAQGVs. The average of the winter (dashed blue line) and the summer (dashed purple line) campaigns is shown.

types, dishwashing frequencies, and cleaning habits. On the other hand, a notable weakness is the relatively small sample size (in absolute terms), preventing the derivation of representative conclusions at either a regional or national level, thereby categorizing this as an exploratory study. Being an exploratory study, another limitation lies in the fact that each restaurant was investigated only once per season, contributing to obtaining a limited database. However, as with all monitoring studies, the present research is aimed at providing an overview of the situation that is generally sufficient to perform a preliminary IAQ assessment. In light of those results, further investigations and interventions could be undertaken should any critical situations be identified. Furthermore, except for the measurements of the hood extraction speed, no ventilation rate data were estimated. The main idea was to use carbon monoxide (CO) as a tracer gas, but several problems with the CO instrument made collecting good-quality data impossible. Moreover, CO_2 could have been used to estimate the ventilation rate after occupants left the kitchen, but we were not allowed to leave the equipment on site after the restaurants closed. At last, the inability to collect simultaneous outdoor samples for VOCs and aldehydes limits the assessment of pollution infiltration dynamics, which would have been valuable for gaining a more complete understanding. Moreover, it is important to emphasize that some pollutants may have strong gradient concentrations over short distances, which makes proximity to monitoring stations a critical factor in interpreting data representativeness.

4. Conclusions

This study provided a comprehensive overview of the concentration level of various gaseous pollutants (inorganic and organic) and UFPs was carried out in 15 restaurant kitchens during cooking and washing activities. To the best of our knowledge, this is the first study of its kind, enabling the investigation of a selected group of indoor air pollutants originating from mixed sources, including cooking and washing/cleaning activities.

Considering the limited monitoring periods and differences in averaging times, the comparison between measured concentrations and guideline limits should be interpreted with caution. However, based on the results and observations, the following conclusions can be drawn:

1. For most of the pollutants investigated, air concentrations were found to be lower than the corresponding IAQGVs, with some exceptions noted for maximum values of O_3 (especially in summer), TVOCs (in both seasons), and NO_2 (especially in winter) measured by direct reading techniques.
2. The indoor air concentrations of benzaldehyde and acrolein exceeded the reference values for several restaurants (in particular, during the hot season).
3. Three cases of exceeding the HCHO threshold value (0.08 ppm) were observed, all within the same restaurant.
4. Quite high concentrations of benzene, up to 14 μg/m^3, were found in winter.
5. Regarding washing and cleaning activities: d-limonene is the most abundant VOC, but the concentrations are lower than the GVs.

In general, higher concentrations of some VOCs (such as d-limonene, α-pinene, and BTEX) were observed in winter than in summer, which could be attributed to several factors, such as changes in ventilation, cooking activities and styles, and microclimatic parameters.

This study confirmed that cooking activities are the main source of indoor pollution inside Italian restaurant kitchens, especially with regards to acrolein and benzaldehyde. However, thanks to the direct-reading instruments, it was demonstrated that the TVOCs average concentrations were higher than the selected IAQGVs, probably due to the combination of cooking and washing activities. Then, in terms of risk management, kitchens should be equipped with correctly designed extractor hoods and the right air exchange rates should be ensured [88]. One suggestion could be the use of low-cost sensors in kitchens for the measurement of major pollutants (e.g., CO_2, HCHO, O_3, and TVOCs) implemented in responsive and adaptive extraction speed of the hoods or, even, ventilation system. Finally, to limit the combination of the emissions from cooking with washing activities, it was suggested to design new kitchens by preferring the layout in which those two activities are carried out in two separate rooms.

A potential future development of this study could involve a specific analysis of the main sources influencing the indoor concentrations of air pollutants in the study sample using a multivariate technique, extracting further key information to identify the most promising risk management options.

Data Availability Statement

The data used to support the findings of this study are included within the supplementary information file(s).

Conflicts of Interest

The authors declare no conflicts of interest.

Author Contributions

Conceptualization: Marta Keller and Andrea Cattaneo. Investigation: Marta Keller. Methodology: Marta Keller, Davide Campagnolo, and Andrea Cattaneo. Data Analysis: Marta Keller and Carlo Dossi. Data curation: Marta Keller. Writing—original draft preparation: Marta Keller, Andrea Cattaneo. Writing—review and editing: Davide Campagnolo, Alessio Carminati, Francesca Borghi, Giacomo Fanti, Sabrina Rovelli, Carolina Zellino, Andrea Spinazzè, Victor G. Mihucz, Carlo Dossi, and Domenico M. Cavallo. Supervision: Andrea Cattaneo, Victor G. Mihucz, Carlo Dossi, and Domenico M. Cavallo.

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Supporting Information

Additional supporting information can be found online in the Supporting Information section. *(Supporting Information)* Additional supporting information including 11 tables, 3 figures and 1 text: Table S1: Checklist used to collect information on the cuisine and activities. Table S2: Detection range and limits of the on-line instrumentation. Table S3: Operating parameters of the ATD-GC-FID instrumentation. Table S4: Operating parameters of the HPLC-UV instrumentation. Text: Identification and quantification of organic molecules. Table S5: List of the investigated analytes and the values of slope (m), intercept (q), correlation coefficients (R^2) and the calculated limits of detection (LODs) and limits of quantification (LOQs), both as mass and concentrations in air. Figure S1: Violin plots of the winter and summer campaigns of indoor pollutants investigated in restaurant kitchens. Results of the Mann–Whitney nonparametric test. Figure S2: Spearman correlation matrix showing relationships between pollutants and other additional parameters (winter campaign). Figure S3: Spearman correlation matrix showing relationships between pollutants and other additional parameters (summer campaign). Table S6: Indoor concentrations of VOCs for the winter and summer campaigns for each investigated restaurant by time-integrated sampling. Table S7: Indoor concentrations of aldehydes for the winter and summer campaigns for each investigated restaurant by time-integrated sampling. Table S8: Guide values of pollutants measured in restaurant kitchens with reference times and sources. Table S9: On-line winter concentrations of indoor pollutants for each investigated restaurant. Table S10: On-line summer concentrations of indoor pollutants for each investigated restaurant. Table S11: Indoor and outdoor concentrations and I/O ratios of benzene, nitrogen dioxide and ozone.

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