EFFECTS OF FRYING ON INDOOR AIR QUALITY

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ABSTRACT

EFFECTS OF FRYING ON INDOOR AIR QUALITY

Frying is an important indoor air pollution source. It may cause chronic health effects on cooks. This study measured indoor air concentrations of volatile organic compounds (VOCs), aldehydes, particulate matter, CO and CO₂ in a small scale restaurant kitchen before, during, and after frying with a margarine produced specifically for frying. Both sampling and monitoring strategies were employed. Individual VOCs, aldehydes, and PM_{2.5} concentrations were determined by sampling. Total VOCs, PM₁₀, CO, and CO₂ concentrations were determined using a monitoring device. Temperature and relative humidity were also monitored as comfort variables in addition to CO₂.

Two campaigns were conducted. In Campaign-1 real working conditions were studied. In Campaign-2 only potatoes were fried with varying amounts. N-heptane, ethyl acetate, nonanal, and n-octane were the realtively higher concentration compounds in both campaigns. The increase in PM_{10} concentrations, however, was much more pronounced: about five times higher when the lowest concentration observed in the very beginning and the peak concentration during frying are compared, and two times higher when the average concentrations are compared. CO and CO_2 concentrations were relatively low, and temperature and relative humidity levels were generally in the comfort zone. The observed PM_{10} concentrations during frying and the average $PM_{2.5}$ concentrations (80-250 μ g/m³) of 4-hr period that covers the all three periods (before, during, and after) in Campaign-1 indicate that chronic health effects are probable for cooks who frequently cook by frying with the frying margarine.

ÖZET

KIZARTMA YAPMANIN BİNA İÇİ HAVA KALİTESİNE ETKİLERİ

Kızartma, aşçılar üzerinde kronik sağlık etkilerine sebep olabilecek önemli iç hava kirliliği kaynaklarından biridir. Bu çalışma, küçük çaplı bir restoran mutfağında kızartma için üretilmiş özel bir margarin kullanımında, kızartma öncesi, kızartma sırasında ve kızartma sonrası oluşan uçucu organik bileşikler (UOB), aldehitler, partikül madde (PM_{2.5}, PM₁₀), karbon monoksit (CO), ve karbon dioksit (CO₂) iç hava kirleticilerinin derişimleri hem örnekleme hem izleme stratejileri ile belirlenmiştir. UOB, aldehitler, ve PM_{2.5} derişimleri örnekleme stratijisi ile, toplam uçucu organik bileşikler, PM₁₀, CO, CO₂ derişimleri ise bir sürekli izleme cihazı ile belirlenmiştir. Ayrıca CO₂'e ek olarak sıcaklık ve bağıl nem seviyeleri konfor değişkenleri olarak izleme cihazı ile izlenmiştir.

Bu çalışma Kampanya-1 ve Kampanya-2 olarak adlandırılan iki kısımdan oluşmaktadır. Kampanya-1'de gerçek işletme koşulları altında, kızartma öncesi, kızartma sırası ve sonrası iç hava kalitesi parametrelerinin derişimleri belirlenmiş, Kampanya-2'de ise sadece farklı miktarlarda patates kızartılarak kızartma öncesi, kızartma sırası ve kızartma sonrası hedeflenen iç hava kalitesi parametrelerinin derişimleri belirlenmiştir. Her iki kampanyada da nispeten yüksek derişimlere sahip UOB'ler n-heptan, etil asetat, nonanal ve n-oktan olduğu tespit edilmiştir. Kızartma önce ve sırası PM₁₀ derişimleri karşılaştırıldığında en yüksek PM₁₀ derişimi kızartma sırasında belirlenmiş ve kızartma öncesi belirlenen en düşük derişimin yaklaşık beş katı, ortalama derişimin ise iki katı olduğu tespit edilmiştir. CO ve CO₂ derişimleri nispeten düşük derişimlerde, sıcaklık ve bağıl nem seviyeleri ise konfor aralığında bulunmuştur. Kampanya-1'de kızartma sırasında gözlemlenen PM₁₀ derişimleri ve 4 saatlık (kızartma öncesi, sırası ve sonrası) ortalama PM_{2.5} derişimleri (80-250 μg/m³) çoğunlukla kızartma margarini ile kızartma yapan aşçılarda kronik sağlık etkilerinin ortaya çıkmasının muhtemel olduğunu göstermektedir.

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CHAPTER 1

INTRODUCTION

Indoor air quality (IAQ) is defined by American Society of Heating, Refrigerating and Air Conditioning Engineers (ASHRAE) (ASHRAE, 1989) as "air in an occupied space towards which a substantial majority of occupants express no dissatisfaction and in which there are not likely to be known contaminants at concentrations leading to exposures that pose a significant health risk". In the modern world, individuals spend nearly 70-90 % of the day time in indoor environments such as houses, work places, schools, public indoor environments (restaurants, cinemas, libraries, shopping centers, stores, etc.). Houses and work places are indoor environments that adult individuals spend time at most (Brasche and Bischof, 2005, Kleepeis et al., 2001). Results of many investigations on indoor air quality show that levels of many indoor air pollutants are higher than outdoor air pollutants levels (Baek et al., 1997; Weschler et al., 2009). Therefore, indoor air quality has greater impact on human health than outdoor air. Commonly faced indoor air pollutants can be grouped under the following catagories: physical pollutants (particulate matter, asbestos, manmade mineral fibres, radon), organic pollutants (Volatile Organic Compounds (VOCs), aldehydes, poliaromatic hydrocompounds (PAHs) and pesticides), inorganic pollutants (carbon dioxide (CO₂), carbon monoxide (CO), nitrogen dioxide (NO₂), sulfur dioxide (SO₂) and ozone (O₃)), environmental tobacco smoke (ETS), and biological agents (dust mites, fungi and bacteria) (Maroni et al., 1995). There are various sources of indoor air pollution. Outdoor air, cooking, smoking, building materials and furnishings, heaters, and office equipments are some of the sources of indoor air pollution (Yocom et al., 1982; Lee et al., 2002). Cooking, in particular frying is one of the most important sources. (Long et al., 2002; Svendsen et al., 2002).

There are many studies about indoor air quality in restaurants, restaurant kitchens, and domestic kitchens in the literature, which will be presented in detail in the next chapter. Some of the studies in literature identify the health effects of indoor air pollutants on human health. Evidences of earlier studies in western countries showed that restaurant cooks had an increased risk of lung cancer as they were continuously

exposed to cooking stove smoke in kitchens (Dubrow and Wegman, 1984; Coggon et al. 1986; Andersson et al., 1997). Cooking in particular frying generates a variety of substances as indoor air pollutants. The mass concentrations and chemical characteristics of the frying emissions depend on various factors which are frying method, food ingredient, oil type, fuel type, stove type, frying duration and frying temperature (Vainiotalo and Matveinen, 1993; Thiebaud et al., 1995; Gertz, 2000; Fortman, et al., 2001; Svendsen et al., 2002; Fullana et al., 2004a,b; See et al., 2006; Zhao, et al., 2007b; Sjaastad et al., 2008; See and Balasubramanian 2008; Katragadda, et al. 2010). Oil type is an important factor which affects the concentrations of indoor air pollutants in frying emissions. In our country vegetable margarine is used for frying in small scale restaurants especially where there is no or insufficient ventilation. Sjaastad et al., (2008) found the highest indoor pollutant concentrations occur while pan frying the food with margarine compared to other oils, and Svendsen et al., (2002) found that frying in a deep fryer in a restaurant kitchen, where ventilation system was insufficient, indoor air pollutant concentrations in frying emissions were high. However there is not any study in the literature that investigated indoor air quality in relation to frying in our country. There is also no study in literature that investigated the effects of deep frying with the special frying margarine on indoor air.

Therefore the overall goal of this research was to determine indoor concentrations of selected air pollutants in a small scale restaurant kitchen in relation to frying with the special frying margarine. Specific objectives of this study were:

- to determine indoor air individual VOC, aldehyde and PM_{2.5} concentrations before frying, during frying, and after frying in the kitchen by active sampling, and
- to measure indoor PM₁₀, TVOC, CO₂ and CO concentrations, temperature and relative humidity levels before frying, during frying, and after frying in the kitchen by using a monitoring device,
- to evaluate if cooks' exposure to indoor air pollution in relation to frying with the special frying margarine is significant and merits further research.

In the following chapters, information regarding indoor air pollutants such as particulate matter ($PM_{2.5}$, PM_{10}), organic pollutants (VOCs, aldehydes), inorganic pollutants (CO_2 , CO) and environmental comfort variables (temperature, relative humidity) and their concentrations related to frying previously reported in the literature

(Chapter 2); materials and methods employed in this study and related quality assurance/quality control measures (Chapter 3); results (Chapter 4); discussion (Chapter 5), and conclusions (Chapter 6) are presented.

CHAPTER 2

LITERATURE REVIEW

2.1. Indoor Air Quality (IAQ)

Individuals spend 90% of their daily times in indoor environments especially houses and workplaces, therefore, indoor air quality has a great impact on human health (Klepeis et al., 2001; Brasche and Bischof, 2005). Various pollutants that are harmful to human health present in indoor environments and many studies have found indoor pollutant levels greater than outdoor levels (Abt et al., 2000; Righi et al., 2002; Rehwagen et al., 2003; Sexton et al., 2004; Rivelino et al., 2006)

There are many sources of indoor air pollution. Various outdoor sources, cooking, smoking, fuel/coal combustion, cleaning, building materials and furnishings, construction materials, heaters, HVAC systems and office equipments are some of the sources of indoor air pollution (Yocom et al., 1982; Lee et al., 2002; Zhang et al., 2003).

People are exposed to various indoor air pollutants depending on the characteristics of the indoor environment and the activity. The concentration of indoor environment activity-related pollutants varies in time depending on the density and duration of the activity. Individuals that are exposed to these indoor air pollutants for a long time are often those most susceptible to the health effects of various indoor air pollutants (Maroni, et al. 1995). Because of the reason that pollutant concentrations can remain in the indoor air for a long time after some indoor activities, health problems may occur on. There are two types of health effects of indoor air pollution. The first one is "Acute Health Effects" which show up after a single exposure or repeated exposures. The second one is "Chronic Health Effects" that show up either years after exposure has occurred or only after long or repeated periods of exposure. Acute health effects include irritation of the eyes, nose, and throat, headaches, dizziness, and fatigue. These acute effects are often short-term and treatable. Symptoms of asthma, hypersensitivity pneumonitis, and humidifier fever, may also show up soon after exposure to some indoor air pollutants. Chronic health effects include respiratory diseases, heart diseases, and cancer which can be debilitating and fatal may show up years after exposure, after long or repeated periods of exposure of indoor air pollutants (U.S. EPA, 2012). Besides of these health problems, different health problems can vary greatly from person to person as a result of exposure to indoor air pollutants.

One of the most important indoor air pollution sources which may cause acute or chronic health problems on individuals is cooking, in particular frying activity. Occupational or non-occupational exposure to emissions from frying includes substantial amounts of airborne particulate matter (PM) that includes ultrafine particles (UFP), PM_{2.5} and PM₁₀ (Ozkaynak et al., 1996 a,b; Brauer et al., 2000; Wallace et al., 2004). In addition to particulate matter a wide variety of organic compounds have been identified in frying emissions. The main volatile compounds which are aldehydes, ketones, alcohols, phenols, alkanes, alkenes, alkanoic acids, carbonyls, poliaromatic hydrocarbons (PAH) and aromatic amines have been identified in frying emissions. Felton, (1995) and also Fortmann et al., (2001) and Kelly, (2001) reported the inorganic pollutants (CO, NO and NO₂) and Lee, et al. (2001) reported CO an CO₂ concentration levels in the emissions from cooking and frying activity.

The following sections offer general information about frying, general information of common indoor air pollutants in frying emissions and major acute and chronic health effects that can occur from exposure to emissions of frying activity and effects of different parameters on frying emissions.

2.1.1. Frying

There are various food preparation processes. One of the mostly used methods is frying, because it is a fast and also appropriate technique for food preparation that gives foods good color and flavor that is appreciated by consumers (Doborgenes et al., 2000). Frying, which is a process of contact food with hot oil and air at high temperatures from 150 °C to 190 °C (Choe et al., 2007), is one of the oldest food preparation processes (Sahin et al., 2009). Different methods of frying are deep-frying, pan frying and stir frying. Deep-frying method is frying by immersing the food in hot oil, frying food in a small amount of oil is called pan frying, frying food in a small amount of oil at high temperature while stirring continuously is called stir frying (See et al., 2008). Deep frying is the frying method that is used domestically and commercially worldwide, which is considered as fast combination of drying and cooking food (Gertz, 2000).

Some of the fried foods are traditional but especially one of the fried foods which is known as "French Fries" is produced with deep frying method has been consumed by people all around the world (Sahin et al., 2009).

Deep frying is a complex operation that involves significant microstructural changes on the surface and in the body of the food and simultaneous heat and mass transfer in opposite directions for water vapor and oil at the surface of the food that is fried (Bouchon et al., 2003). Chemical reactions which occur in frying oil with contact among oil, food and air are hydrolysis, oxidation, and polymerization as a result of which, volatile or nonvolatile compounds are formed in oil. Volatile organic compounds evaporate in to the air with steam mostly while a portion of them remain in food. (Choe et al., 2007; Meesuk and Vorasith, 2006).

2.2 Pollutants from Frying Emissions

2.2.1. Organic Pollutants

In indoor environment, there are wide variety of known organic chemicals. Organic pollutants, can be divided according to their chemical character as aldehydes, ketones, aliphatic hydrocarbons, aromatic hydrocarbons, alkanes, etc., their pysical properties as boiling point, vapour pressure, carbon numbers etc., and according to their potential health effects as irritants, neurotoxics, carcinogens, etc. (Molhave et al., 1997).

World Health Organization (WHO) made the Volatile Organic Compound classification according to a wide boiling-point range. The boiling point ranges from <0 °C to 50 °C-100 °C are known as Very Volatile Organic Compounds (VVOC), boling point of 50 °C-100 °C to 240 °C-260 °C are known as Volatile Organic Compounds (VOC) and between the boiling point 250 °C -260 °C to 380 °C - 400 °C are known as Semi-Volatile Organic Compounds (SVOC). All classes of VOCs are typically sampled by adsorption on carbon molecular black, charcoal or the porous polymer Tenax. Over 900 different VOCs had been detected in the indoor air (U.S. EPA, 1989). Various studies of indoor air quality have determined more than 250 organic compounds that exceed 1 ppb (Namiesnik et al., 1992).

In the study of Lee et al., (2001) emissions from different styles of cooking activities were determined. This study was performed in four restaurants which were a

Korean Barbecue restaurant, a Chinese hot pot restaurant, a Chinese dim sum restaurant and a Western canteen. In Korean Barbecue restaurant where the food was fried in a pan in peanut oil on a LPG gas stove, the highest VOC concentrations were determined. Total hydrocarbon (THC) concentrations ranged from 9.7 µg/m³ to 14.2 µg/m³. The mean THC concentration was determined as 11.4 µg/m³. Specific determined VOCs were benzene, toluene, methylene chloride and chloroform for which the mean concentrations were found as 18.4 µg/m³, 156 µg/m³, 0.6 µg/m³ and 14.9 µg/m³ respectively. Another study was performed in Norway in nineteen restaurants' kitchens (four hotel kitchen, two hamburger chain restaurant, ten a la carte restaurant with grill and three small local restaurant mostly serving fried food) were selected as sampling locations. All nineteen restaurants had deep fryers that were used for patatoes frying, equipped with ventilation hoods. Temperature of oil was ranged from 160 °C to 190 °C. Oil types that were used for frying were soy oil, sunflower oil and palm oil. The highest aldehyde concentrations were determined at the breathing zone of the cook in small local restaurant kitchen that were serving mostly fried food and had a insufficient ventilation system. The mean concentrations of formaldehyde, acetaldehyde and total aldehydes were found as 14 $\mu g/m^3$, 102 $\mu g/m^3$ and 119 $\mu g/m^3$ respectively (Svendsen et al., 2002).

2.2.2. Particulate Matter (PM)

PM represents a complex mixture of organic and inorganic substances. Mass and composition can be divided into two groups which are coarse particles that are larger than 2.5 μ m (2.5 μ m-100 μ m) in aerodynamic diameter and fine particles smaller than 2.5 μ m (0.0001 μ m- < 2.5 μ m) in aerodynamic diameter. Special focus is currently paid to PM_{2.5} which is the respirable particle size and such particles can cause damage to human health (Maroni et al., 1995)

PM₁₀ concentration levels have been measured in several studies during various cooking procedures (Abt et al., 2000a; Kelly, 2001; Lee et al., 2001; Fortmann et al., 2001; Lee et al., 2007). In addition, Fortmann et al., 2001; He et al., 2004; Evans et al., 2008 which reported that PM_{2.5} concentrations increases by thirty times during frying in domestic kitchens.

 $PM_{2.5}$ concentrations were found in the range of 24–201 $\mu g/m^3$ in residential kitchens during frying, with peak concentrations >400 $\mu g/m^3$ (Brauer et al., 2000). In

Fortmann et al., (2001) study various types of food (bacon, tortillas, fish, beef,pork roast and popcorn) were cooked with different cooking methods. One of them was french fries that fried in a deep fryer in the uses of vegetable oil. Four sampling location were selected one of them was kitchen. Samplings were performed at the breathing zone of the cook and also a monitoring device was used for continuous monitoring of particulate matter. The temperature of the deep fryer was set to 190°C and the quantity of the one bag of French fries was 3045g when one bag was cooked, second one was fried. Two types of stoves that were gas and electric stove were used. PM_{2.5} concentrations which were determined at the breathing zone of the cook were 374 μg/m³ and 195 μg/m³ for electric and gas stove applications respectively. PM_{2.5} concentrations in the kitchen air were determined as 66 µg/m³ 116 µg/m³ for electric and gas stove applications respectively. It can be seen from the results that sampling PM_{2.5} concentrations that were determined at the breathing zone of the cook were higher than the PM_{2.5} concentrations that were determined in the kitchen indoor air during frying period. Average estimated PM_{2.5} concentrations in total exposure period were determined as 123 μg/m³, and 128 μg/m³ when electric stove and gas stove were used respectively. PM₁₀ results which were determined at the breathing zone of the cook during frying were 207 µg/m³ and 342 µg/m³ for electric and gas stove applications, respectively. Average PM₁₀ concentrations during frying measurement results were determined as 724 μg/m³ 850 μg/m³ in indoor air of kitchen and average PM₁₀ concentrations were found as 715 µg/m³ 534 µg/m³ in total exposure period for electric and gas stove applications respectively.

Lee et al., (2001) investigated the indoor air concentration levels of indoor air pollutants that were emitted from cooking activity in different styles of restaurants which were a Korean barbeque restaurant, a Chinese dim sum restaurant, a Chinese hot pot restaurant and a Western cafeteria. The highest average indoor $PM_{2.5}$ and PM_{10} concentration levels were found as 1167 $\mu g/m^3$ 1442 $\mu g/m^3$, respectively in Korean barbeque restaurant where the food was fried in a pan.

 $PM_{2.5}$ and submicrometer particle concentrations were determined in kitchens during cooking activities (frying, grilling, toasting) in fifteen houses in Australia (He et al., 2004). The study was performed at two meters distance from the stove. In two of the houses there were gas stoves and rest of the houses had electric stoves. Median peak $PM_{2.5}$ concentration level was reported as 745 μ g/m³ during frying activity.

Kelly, (2001) determined $PM_{2.5}$ and PM_{10} concentrations in a test chamber (17.5m³) some time after cooking finished and under realistic conditions in a research house during cooking activities. The repetitions of various cooking activities were conducted on both a gas and an electric stove. The highest indoor $PM_{2.5}$ and PM_{10} concentration levels occurred in the kitchen of the research house at the breathing zone of the cook during frying of hamburgers while the lowest concentrations occured in the living room or bedroom. Frying hamburgers on the gas stove and the electric stove produced average $PM_{2.5}$ concentrations of 115 μ g/m³ and 230 μ g/m³ and PM_{10} concentrations of 118 μ g/m³ and 328 μ g/m³, respectively in the research house kitchen at the breathing zone of the cook. In the test chamber frying of hamburgers were registered as high emitting processes in terms of particulate emissions.

 $PM_{2.5}$ concentrations were determined by See and Balasubramanian, (2008) during steaming, boiling, stir-frying, pan frying and deep frying of one pack of tofu. The background concentration was determined as 15.4 $\mu g/m^3$. The average $PM_{2.5}$ concentrations were found as 72.3 $\mu g/m^3$ during steaming, 91.6 $\mu g/m^3$ during boiling, 120 $\mu g/m^3$ during stir-frying, 130 $\mu g/m^3$ during pan-frying and 209 $\mu g/m^3$ during deepfrying.

2.2.3. Inorganic Pollutants

A wide range of inorganic pollutants which affetcts the indoor air quality, can be emitted by indoor environment appliances, which includes carbon dioxide (CO₂), carbon monoxide (CO), nitrogen dioxide (NO₂), sulfur dioxide (SO₂) as the main pollutants.

Carbon dioxide (CO₂) is a colourless, odourless gas that is the main combustion product of cooking and heating purposes. Some health problems occur as a result of CO₂ exposure, which may be fatal at extreme concentrations. Respiration is affected and breathing becomes faster and more difficult concentrations above 1.5%. Concentrations above 3% can cause headaches, dizziness and nausea and concentrations above 6-8% can result in stupor and death (Maroni et al., 1995).

Carbon monoxide (CO) is an odorless, colorless and toxic gas which is the product of incomplete combustion of carbon-containing materials. Exposure to CO at low level, this gas causes mild effects that are often mistaken for the flu. The symptoms

which can vary from person to person depending on age, health, exposure concentration and length of exposure are headaches, dizziness, disorientation, nausea and fatigue (U.S. EPA, 2012).

There are numerious different nitrogen oxides (NO_x) in air, among which nitrogen dioxide (NO₂) is the most widely considered in indoor air pollution studies because of NO₂ is a reactive gas, and poses the greater health hazard (Fisk et al., 1987). NO₂ is a water soluble red to brown gas with a pungent, acrid odor that is produced during high temperature combustion from the combination of nitrogen and oxygen from air. Indoor sources of NO₂ include environmental tobacco smoke (ETS), gas appliances, kerosene heaters, and fireplaces (Maroni et al., 1995). Health effects of NO₂ exposure can vary according to exposure concentration levels. Concentrations at 0.5 ppm have been shown to cause effects in people with asthma (Cooper et al., 2002). Exposure to about 5 ppm concentration of NO₂ can cause respiratory distress, concentrations of about 50 ppm can cause chronic lung disease and exposure greater than 150 ppm concentration level can be fatal (Fisk et al., 1987).

Sulfur dioxide (SO_2) is a colourless gas with a strong pungent odour which can be detected by the human nose at about 0.5 ppm. It is readily soluble in water and can be oxidized within airborne water droplets. Health effects from exposure to SO_2 include sensitivities of the respiratory tract for short-term exposure to increased risk of chronic bronchitis with long-term exposure (Maroni et al., 1995).

There are some studies about inorganic pollutants that occured during frying activity. In the study of Kelly (2001) emissions of PM, NO_X an CO in an experimental chamber (17.5 m³) during pan frying and six other different types of cooking activity were determined. Another study of cooking emissions was performed by Fortmann et al., (2001). In the study PM_{2.5} and PM₁₀ particles, carbon monoxide (CO), nitrogen oxide (NO) and nitrogen dioxide (NO₂) were measured during different types of frying activity including deep frying of french fries. Another study was conducted by Lee et al., (2001), in which CO and CO₂ concentrations were determined as inorganic pollutants in restaurants during different styles of cooking activities. In Korean barbecue restaurant where the food was fried in a frying pan in peanut oil on LPG gas stove, CO concentrations ranged from 11543 μ g/m³ to 18743 μ g/m³ with a mean concentration of 15703 μ g/m³. CO₂ concentrations ranged from 1364 μ g/m³ to 1868 μ g/m³ with a mean of 1648 μ g/m³.

2.3. Health Effects of Exposure to the Chemical Components in Frying Emissions

Emissions from cooking especially from frying generate several specific indoor air pollutants, neither occupational nor non-occupational exposure to these emissions from cooking or frying may give rise to adverse health effects. International Agency for Research on Cancer (IARC) have classified emissions from high temperature frying as 'probably carcinogenic to humans (Group 2A)' (IARC, 2006). Studies have shown that emissions from cooking contain aldehydes (Vainiotalo and Matveinen, 1993; Svendsen et al., 2002). Svendsen et al., (2003) stated that inhalation of aldehyde emissions irritate to the airways. There are some epidemiological studies which were indicated that exposure to emissions of cooking oil may be important for lung cancer for non smoking Chinese females, because of a traditional reason. Females prepare the meals for their family in China and Chinese style cooking includes deep-frying of food in oil and stir frying (Wu-Willans et al., 1990). Also similar study was performed in Shangai where Chinese rapeseed oil was used for preparetion of meal by women. Zhong et al., (1999) found in this study that higher lung cancer risk has been found among women who most often cook with rapeseed oil. Heating the oil generates volatiles which are complex mixture of aldehydes and nonvolatile products because of thermal and oxidative decomposition of the oil. These complex mixture of aldehydes has been considered intermediaries of oxidative stress which may play some role in the mechanisms of tumor promotion in human body (Trush and Kensler, 1991). Coggon et al., (1986) also reported that there was an increased risk of respiratory tract cancer in cooks and bakers.

2.4. Effects of Different Parameters on Frying Emissions

Cooking in particular frying generates a variety of substances as indoor air pollutants. The mass concentrations and chemical characteristics of the frying emissions depend on various factors which are frying method, food ingredient, oil type, fuel type, stove type, frying duration and frying temperature (Vainiotalo and Matveinen, 1993; Thiebaud et al., 1995; Gertz, 2000; Fortman et al., 2001; Svendsen et al., 2002; Fullana

et al., 2004a,b; See et al., 2006; Zhao et al., 2007; Sjaastad et al., 2008; See and Balasubramanian, 2008; Katragadda et al., 2010).

Fortmann et al., (2001) studied effects of different food types, different frying types, and different stove types on the concentrations of frying emissions. PM (PM_{2.5} and PM₁₀₎, carbon monoxide, nitrogen oxide, nitrogen dioxide, PAHs and aldehydes were measured. The frying activities were studied under standard conditions and using two stove types which were electric stove and gas stove. Cooking activities included wok stir-frying of chicken and vegetables, deep frying of french fries and pan-frying of bacon, tortillas or hamburgers under standard conditions. Wok stir-frying was performed with peanut oil at high temperatures, using chicken and vegetables as food. Deep frying method was used for frying french fries. Results showed that PM_{2.5} concentrations were higher when electric stove was used. Also Fullana et al., (2004a,b) study, it was found that the generation rates of aldehydes were dependent on heating temperature level, and generation rates of aldehydes increased with increases in temperature. The emissions of low molecular weight aldehydes from deep frying of extra virgin olive oil and olive oil were similar and were lower than the levels of aldehydes that observed from canola oil under similar conditions. In a study by Sjaastad et.al, (2008), four different types of oil were used for frying under real life conditions. Results showed that during frying with margarine in a pan gave statistically significantly higher levels of aldehydes and particulate matter than frying with other three different kinds of oil which were rapeseed oil, soybean oil and virgin olive oil. In the study of See and Balasubramanian, (2008) it was demonstrated that method of frying is an important factor for indoor air pollutant concentration levels. In the study three different types of frying methods that were deep frying, stir frying and pan frying were used to fry plain tofu. The sampling location was 1.5 m above the ground and close to the breathing zone of the cook. The results showed that the deep-frying method emitted the highest concentrations of particulate matter and most chemical components that were observed. Katragadda et al., (2010) indicated that the temperature had an impact on the volatile organic compounds including aldehydes. During heating four different oils to different temperature levels, the same volatile compounds were present in emissions at different levels of temperatures, but the quantities of volatile organic compounds increased significantly when the temperature was risen.

CHAPTER 3

MATERIALS AND METHODS

Sampling and analytical procedures and quality control and assurance measures for 48 VOCs, 14 aldehydes and particulate matter ($PM_{2.5}$) are described in this chapter. In addition, measurement of particulate matter (PM_{10}), TVOC, CO and CO₂ indoor air concentrations and temperature and relative humidity levels are described.

This study included two main parts:

- 1. Sampling and Measurement
 - a. Sampling of VOCs, aldehydes and PM_{2.5} in a small-scale restaurant kitchen (Field Study)
 - b. Monitoring of PM₁₀, TVOC, CO₂, CO, temperature and relative humidity in a small-scale restaurant kitchen (Field Study)
- 2. The analyses of the samples for the concentration determination (Laboratory Study)

3.1 FIELD STUDY

3.1.1 Restaurant Selection

Generally small scale restaurants use margarine for frying. In addition small scale restaurants are more appropriate to characterize the work environment of the cooks as the owners tend to be more willing to participate. Therefore a small scale restaurant located in Urla, İzmir was selected based on frying oil, willingness to participate the study and accessibility.

Restaurant Kitchen: This restaurant was serving mostly fried food using a deep fryer. The deep fryer had two parts and each part had 5 liter capacity. However, only one part of deep-fryer was used. Mostly frozen ready to cook french cut potatoes were cooked in the deep-fryer. Other usage of deep-fryer was to fry nuggets, schnitzels, hamburger patties and chicken patties. The temperature of the deep fryer was ranged

from 160 to 190 °C. The oil used was a kind of special margarine for frying called 'Vegetable Margarine' or 'Vegetable Dehydrated Oil' which contains refined vegetable oils (palm oil). In this restaurant kitchen, there was no hooded exhaust system thus cooking fumes from frying disposed in the volume. Windows were kept closed all the time while the kitchen door was kept open.

3.1.2. Frying Procedure

3 liters of vegetable margarine was added to one part of deep fryer that had a 5 L capacity. Vegetable margarine was not renewed daily in this small scale restaurant kitchen. When it decreased to minumum level of the deep fryer, it was renewed. Therefore it was depended on the quantity of the order. On average, the frying oil turnover time was four or five days in this restaurant. On stand by deep fryer was kept at 50 °C. When an order came deep fryer was heated up to 160-180 °C with the temperature setting button on the fryer. After heating up to required temperature, frozen product was added to deep fryer. Depending on the product, frying durations and temperatures varied. In Table 3.1 frying temperatures and durations are shown for different frozen products. Frozen products were kept in the freezers at -18 °C.

Table 3.1. Frying Temperatures and Frying Durations

	<u> </u>	0
Frozen Product	Frying Temperature (°C)	Frying Duration (min)
French Fries	180	4-5
Hamburger Patties	160	5
Chicken Patties	160	5
Schinitzel	160	5
Nuggets	160	5

3.1.3. Sampling and Monitoring

Harvard Impactor: Harvard sampler is used to sample airborne particles with different aerodynamic size PM_{2.5} and PM₁₀. In Figure 3.1 diagram of Harvard Impactor is shown. The impactor consists of filter holder base and after filter holder body with filter hold down clamps, nozzles, sampling inlet, after filter in 2" x 2" filter slide and impaction plates. Standard Anderson type dichotomous sampler filter holder rings were

used in this study. Flow of the Impactor is ranged from 1 to 30 l/min. In this study, only $PM_{2.5}$ concentrations were collected using a pump connected to the Harvard Impactor with a flow rate of 20 l/min.

SKC Air Check 2000 Pump: A SKC AirChek 2000 Pump an advanced programmable sample pump that accurately and directly measures flow with Nickel Cadmium (NiCad) battery was used for collecting VOC and aldehyde samples. Flow of the pump is ranged from 1000 to 3250 ml/min. In addition to this using a low flow adapter kit sampling can be performed in range between 5-500 ml/min. In this study two SKC Air Check 2000 pumps were used. One of them was used for aldehyde sampling at the high flow rate range, other one was used at low flow rate range for VOC sampling.

Calibration Device: Calibration devices were used to calibrate the pumps (SKC Air Check 2000 pumps and Harvard Impactor). High flow rate sampling pumps were calibrated before and after sampling with a flow controller called Bios International Corp, Defender 510-H, low flow rate sampling pumps were calibrated with Bios International Corp, Defender 510-L.

Tenax TA Sorbent Sample Tube: Tenax TA is a traditional sorbent (porous polymer) for trapping of volatiles and semi-volatiles from air based on 2.6-diphenylene oxide. Its unique structure provides alternate and desirable adsorption characteristics compared to other porous polymers. Tenax TA is suitable for use in EPA Method TO-17 or IP-6A and other thermal desorption applications. Using thermal desorption techniques, detection of volatile organics in the ppb and ppt levels is feasible. Tenax TA (SKC 226-340) 1/4" OD x 3 1/2" long and contains 100 mg sorbent was used for VOC sampling.

DNPH Coated Silica Gel Sorbent Tube: Commercially available formaldehyde sample tube (SKC. 226-119) is 6-mm OD x 110-mm long and contains a 300-mg front sorbent section and a 150-mg backup sorbent section. The sorbent is silica gel with ultra-low background coated with 2.4-dinitrophenylhydrazine (DNPH). The DNPH chemistry lends itself to monitoring formaldehyde and other aldehydes simultaneously in the workplace environment.

Sorbent Tube Accesorries: Single / Dual Adjustable Low Flow Holder is designed to be used for higher flow pumps for low flow applications from 5 to 500 ml/min using sorbent sample tubes. In this study Single Adjustable Low Flow Holder was used to sample VOC concentrations and Dual Adjustable Low Flow Holder

duplicate tube sampling of VOC concentrations. Flow rate can be adjusted using the convenient flow adjust screw on the holder.

Precision Balance: A precision balance (Sartorius CPA225D) which has a readibility range from 0.01mg to 0.1 mg was used to weigh the 37-mm glass fiber filters before sampling and after sampling.

Air Monitoring Standards: Japanese Indoor Air Standard Mix (100 μ g/mL in methanol: water (19:1), 52 Chemicals, Supelco 4M7537-U) was used for GC/MS calibration and Aldehyde / Ketone DNPH TO11 / IP6A Mix (15 μ g/mL each component in acetonitrile (aldehyde equivalent), Supelco 4M7285-U was used HPLC calibration in this study.

Filters: 37-mm Glass Fiber Filters (Pall, Type A/E, 1μm pore) were used to collect airborne particles (PM_{2.5}) with Harvard Impactor Device. A Desiccator was used to protect the 37-mm glass fiber filters from humidity.

3M Quest Technologies EVM-7: The 3 M EVM-7 device monitors both particulates and air quality in one compact instrument. It is a portable area monitoring instrument that measures and stores concentration levels of particulate matter one of the following at a time (PM_{2.5}, PM₄, PM₁₀ and PM), TVOC, CO₂, CO, temperature, relative humidity and dew point concurrently. This monitoring device measures PM concentrations with using a 90° optical light emitting photometer. TVOC concentrations are recorded with a photo ionization detector (PID sensor) in ppb units. With a nondispersive infrared sensor (NDIR) CO₂ and CO concentrations are recorded in ppm units. The flow rate of this monitoring device is 1.67 L/min.

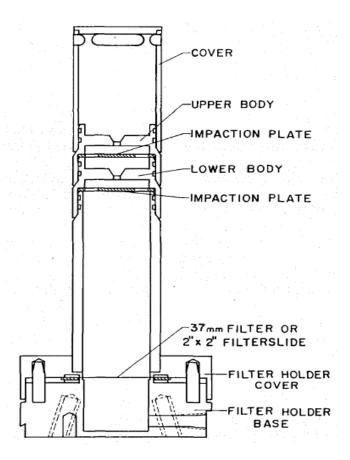


Figure 3.1. Diagram of the Harvard Impactor (Source: Marple et al., 1997)

3.1.4. Sampling and Measurement Program

Sampling and measurement program consists of two campaigns. A three day sampling/measurement program was conducted in each campaign. In the first campaign, fried products were mostly french fries but also nuggets, schnitzels, hamburger patties and chicken burger patties were also fried depending on customer orders. In the second campaign only french fries were served. The amount of french fries increased from 1250 g on the first day to 2500 g on the second day to 3750 g on the third day. First campaign was in November 2012. The second campaign was in February 2013. Because the restaurant served university students and the personnel, orders mainly came on lunch time. Therefore, all the sampling was performed in weekdays from 11.00 to 15.00. Each day was diveded into three sampling periods for VOC and aldehydes. *'Before Frying'* samples were collected from 11.00 to 12.00 to determine background levels. *'During Frying'* and *'After Frying'* periods were 12.30 to 13.30 and 14.00 to 15.00 respectively. The PM_{2.5} sampling duration was set to four hours (240 min) due to

concern about collecting sufficient amount of particulate matter for weighning. PM₁₀, TVOC, CO₂, CO concentrations were measured, temperature and relative humidity levels were monitored with EVM-7 continuously throughout four hour (240 min) period from 11.⁰⁰ to 15.⁰⁰.

3.1.5. VOCs and Aldehyde Sampling

VOCs and Aldehyde sampling time and sampling volumes were determined based on U.S. EPA methods. U.S. EPA Compendium Method TO-17 (Determination of Volatile Organic Compounds in Ambient Air Using Active Sampling Onto Sorbent Tubes) (U.S. EPA, 1999) which describes a sorbent tube sampling and analysis with thermal desorption/gas chromatography method for VOCs in air presented convenient sample volumes as 1 and 4 liter in a sampling time of 5-60 minutes. The adequite sampling flow rate range is 10 to 200 ml/min. In this study, the sampling flow rate for VOCs sampling was determined as 66.7 ml/min for an hour collecting 4 liters of air. Tenax TA sorbent tubes are suitable trapping mediums for using in EPA Method TO-17 therefore, Tenax TA sorbent tubes were utilized for VOCs in this study. EPA Method IP-6A (Determination of Formaldehyde and Other Aldehydes in Indoor Air) (U.S. EPA, 1990) which describes solid adsorbent sampling followed by high performance liquid chromatographic analysis (HPLC) for aldehydes in indoor air, presented appropriate flow rates of 100-1500 ml/min for short term sampling times of 5-60 minutes and long term sampling time 1-24 hours. The flow rate and sampling time were set as 1000 ml/min and 60 minutes, respectively. In this study, aldehyde samples were collected on DNPH-coated silica gel sorbent tubes because they meet specifications of Environmental Protection Agency (EPA), and Occupational Safety and Health Agency (OSHA) methods.

The sampling flow rates were measured before starting sampling and end of each sampling period with flow calibrators. High flow rate pumps were calibrated with Bios International Corp, Defender 510-H (500-30000 L/min) and low flow rate pumps were calibrated with Bios International Corp, Defender 510-L (1-500 L/min). Flow calibrators were connected to the pumps with tygon tubing. One of the DNPH silica gel sorbent tubes and one of the Tenax TA tubes were assigned as calibration tubes.

Difference between flow rates before and after sampling was ± 1 ml/min for VOCs and for aldehydes sampling ± 15 ml/min.

In EPA Method TO-17 it is emphasized that before first use of Tenax TA tubes for VOC sampling they must be thermally conditioned with at least 50 ml/min of pure Hellium carrier gas at the temperature of 350 °C for newly packed tubes at least 2 hours and 30 minutes for pre conditioned, purchased tubes to remove residual components. In this study, before first use of Tenax TA tubes, they were reconditioned at 300 °C for 1 hour. On subsequent uses, Tenax TA tubes will not require conditioning as before first use. Instructions in EPA Method TO-17 was applied before subsequent uses, Tenax TA tubes were conditioned at 250 °C, purge time was 1 minute and desorb time was 3 minutes. Tenax TA tubes were sealed with ungreased Swagelok®-type, metal screwcaps and combined PTFE ferrules. The screw caps should be tightened by hand and then an extra 1/4 turn with a wrench and they were wrapped with aluminum foil individually to protect from the light, after that they were placed in a clean, airtight, opaque storoge container with ice packs to maintain at 4 °C during transportation to sampling location. DNPH silica gel tubes were single-use tubes that did not require pre-conditioning. DNPH silica gel sorbent tubes were kept in the freezer until sampling and transported with same storage container with Tenax TA tubes to maintain at 4 °C and protect from the light.

At sampling location, fistly, pumps were calibrated. Sampling tubes were kept in their storage container during the calibration of the pumps. After that, with clean gloves the one of the Tenax TA tubes and one of the DNPH silica gel tubes were removed from storage container and waited for a while to equilibrate the tubes temperature with indoor air temperature. Other remaining two Tenax TA tubes and two DNPH silica gel tubes were placed in a portable refrigerator until sampling.

Firstly, for 'before frying' sampling Tenax TA and DNPH silica gel tubes were prepared. Aluminium foil and Swagelok®-type fittings and PTFE ferrules of Tenax TA tube was removed and it was attached to the non-outgassing flexible tube that was attached to adjustable low flow holder which was connected to the SKC Air Check 2000 pump with tygon tubing. Because of Tenax TA tubes are bidirectional, they were attached to pump in the direction of the arrow that was on the Tenax TA tube. DNPH silica gel sorbent tubes edges were cutted with a glass cutter and was attached to the tube holder which was attached to the pump with a tygon tubing. In figure 3.2 DNPH Silica gel sorbent tube is shown. Indoor air sampling and measurement equipments were

placed approximately 1.5 meters above the ground at breathing zone height and near the deep fryer. All sampling and measurement equipments were started at the same time. After sampling, with Swagelok®-type fittings and PTFE ferrules, Tenax TA tubes were sealed and they were wrapped in aluminum foil. DNPH Silica gel tubes were capped with sealed caps and they were also wrapped in aluminum foil. All sample tubes were labeled for identification and were placed in the refrigerator. Also sample date and time, sample location were recorded. After that other sample tubes were removed from refrigerator for 'during frying' sampling and 'after frying' sampling all these steps were applied respectively. At the end of the sampling period the flow rate of the pumps were checked with the flow calibrater. All flow rates measured at the end of sampling agreed within 10% with that measured at the start of the sampling period as in stated in EPA Method TO-17. Then all the sample tubes were removed from the refrigerator and placed to the refrigerated storage container for transport to the laboratory. The tubes were stored at < 4 °C in the refrigerator until analysis. The time between sampling and analysis should not exceed 30 days as stated in the EPA Method IP-6A and EPA Method TO-17. In this study, the Tenax TA tubes were analyzed in two days. DNPH silica gel tubes were analyzed in 21 days.

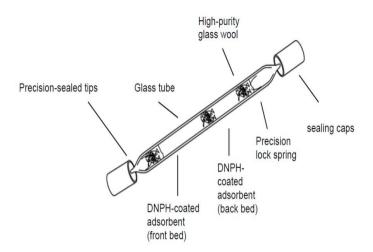


Figure 3.2. DNPH Coated Silica Gel Sorbent Tube (Source: SKC, 2004)

3.1.6. PM_{2.5} Sample Collection

PM_{2.5} samples were collected with an Harvard impactor connecting to a sampling pump. In this study, the flow rate of the pump was set to 20 L/min. This pump was also calibrated with Bios International Corp, Defender 510-H (500-30L/min) before sampling and at the end of the sampling.

Before first use of 'impaction plate' which is one of the parts of Harvard Impactor was placed in a beaker containing Alconox and hot water and soaked for 30 minutes, agitating periodically for cleaning. It was rinsed with warm water and washed a second time. After that, the plate was rinsed again in clean beaker. The beaker was placed in a sink with tap running, the water was let to fill the beaker and overflow. The plate was drained and dried in an oven at 60 °C for 3 hours. After the plate cooled, it was placed in zip-lock bag.

PM_{2.5} samples were collected on 37-mm glass fiber filters. Before sampling, filters were wrapped loosely with aluminum foil and baked overnight at 450 °C in a muffle furnace to remove any organic residue. They were then placed in a desiccator to protect from moisture at least 24 hours. After then, they were weighed with a precision balance Sartorius CPA225D with 0.01 mg accuracy. Each filter was weighed for three times with precision balance. Then each of the weighed filters was placed in storage containers and all filters were labeled for identification. Until sampling, filters were kept in the desiccator. Storage containers of the filters were sealed with parafilm and were wrapped in aluminum foil for transporting to the sampling location.

At sampling location, filter was removed from storage container and aluminum foil and parafilm were unwrapped. Filter was placed in the sampler filter holder rings with the help of a tweezer. The impaction plate of the pump require greasing before each sampling therefore, upper surface of the impaction plate was greased. After that, all parts of the Harvard impactor were assembled except for the sample cover. Instead of sample cover, calibration cover was placed. With tygon tubing it was attached to the flow calibrator for calibration. After calibration, the calibration cover was removed and sample cover was placed. At the end of the sampling, calibration steps were repeated. Filter was removed with the tweezer and was placed in storage container were sealed with parafilm and wrapped in aluminum foil for transportation to the laboratory. After

filters were stored in the dessicator at least 24 hours, they were weighed for three times with the precision balance.

3.1.7. Measurement of Indoor Air Pollutants with an Air Monitoring Device

3M Quest Technologies EVM-7 was used for measurement of indoor PM_{10} , TVOC, CO_2 and CO concentrations, monitoring temperature and relative humidity levels. Flow rate of EVM-7 is constant at 1.67 l/min. This monitoring device measures PM_{10} concentrations using a 90° optical light emitting photometer. TVOC concentrations are measured with a photo ionization detector (PID sensor) in ppb units. With a nondispersive infrared sensor (NDIR) CO_2 and CO concentrations are recorded in ppm units.

All sensors of the mointoring device were factory calibrated but before first use of the pump in the run mode for particulate matter measurement, zero filter calibration is recommended. Before zero calibration, impactor must be greased with silicon grease and this application must be repeated periodically depending on the density of air particles floating in the intended measurement environment. After greasing and cleaning impactors, zero calibration was performed. Impactor was set to PM and Zero/Hepa Filter was inserted into the impactor. In the calibration screen, when the level stabilized zero calibration was ended and the level was saved. At measurement location, EVM-7 was placed approximately 1.5 meters above the ground at breathing zone height and near the deep fryer. Concentrations of PM₁₀, TVOC, CO₂, CO and levels of temperature and relative humidity were recorded in every 15 seconds as minumum level, maximum level, average level and short term exposure level (STEL). After sampling, all recorded data was transferred to the computer.

3.2. Laboratory Study

In this part, laboratory study which includes the preparation of the calibration solution standards analyses and Quality Assurance/Quality Control (QA/QC) measures are described for VOCs and aldehydes. Analysis of VOC and operating conditions of Thermal Desorption - Gas Chromatography-Mass spectroscopy (TD-GC-MS) are presented. Analysis of aldehydes, extraction of analytes from DNPH silica gel tubes and operating conditions of High Performance Liquid Chromatograph (HPLC) are described. Finally, QA/QC measures including field blanks concentrations, duplicate precisions, and limit of quantifications (LOQ) were presented.

3.2.1. VOC Analysis

Tenax TA tubes that were used for VOCs sampling were thermally desorbed with TD-GC-MS system. Thermal desorption is a physical separation process that transfers contaminants from one phase to another; in this case of organic compounds from solid collection medium by heating to a temperature that organic contaminants volitilize. Air, combustion gas, or inert gas can be used as transfer medium for vaporized components (Pal et al., 1998). In this study, Unity Thermal Desorption System (Markes International Ltd) which was compatible wih injector/septum area of the gas chromatography/mass spectrometer (Agilent Technologies 6890N) was used for thermodesorption at 250 °C. Helium, was used as a carrier gas (inert gas) at 1.1 ml/min to transfer the vaporized volatile organic compounds in the thermal desorption system. Before thermal desorption, some procedures must be followed as it was stated in EPA Method TO -17. The following sample and system integrity checks and procedures were carried out automatically before thermal desorption:

Tube Leak Testing: Each tube must be stringently leak tested at the GC carrier gas pressure, without heat or gas flow applied, before analysis. Tubes which fail the leak test should not be analyzed.

Sample Flow Path Leak Testing: All parts of the sample flow path should be stringently leak tested before each analysis without heat or gas flow applied to the sample tube. These measures prevent a leak in the system which can cause loss of

sample and incorrect flows through the tube trap and down the column, causing loss of sensitivity and reproducibility.

Purge Air: Purge air from the tube and sample flow path at ambient temperature using carrier gas immediately before tube desorption. It helps to dry the sample and prevents analyte and sorbent oxidation so minimizing artifact formation, ensuring data quality and extending tube lifetimes. In this study, with clean gloves, Swagelok®-type fittings were removed from Tenax TA tubes and the tubes were purged with a forward (sampling direction) flow of, 50 ml/min of Helium for 1 minute.

In the laboratory, for analysis one of sample collected Tenax TA tubes, was removed from the refrigerated storage container and Swagelok®-type fittings and PTFE ferrules were removed. The Tenax TA tube was placed on the thermal desorber. Firstly, sample tube was heated with carrier gas flowing through it, at the same time the sample was desorbed from the tube at 250 °C to achieve the transfer of VOCs to the cold trap at -10 °C during 3 minutes. Thermal desorption continued until all target analytes were transferred to the focusing trap. Typically, the cold trap sorbent is the same as in the sample tube, but it is a much smaller sorbent bed a maximum of 60 mm long and only 2 mm diameter. After desorption completed, focusing trap was heated rapidly and a reverse flow of pure helium carrier gas was applied. Analytes were transferred to the column in a narrow band of vapor that enhanced separation and resolution of the compounds. The GC run was initiated based on a time delay which was 8 minutes after the start of thermal desorption. In this study, GC was equipped with HP-VOC 60 m. 320 µm. 1.80 µm column. The operating conditions of thermal desorption system is summarized in Table 3.2. The run time for GC analysis, was 41.25 min. GC-MS operating parameters are summarized in Table 3.3, and an example gas chromatogram is shown in Figure 3.3. Chemical properties of each VOC compound was identified using molecular weight (M.W), boiling point/range, target ions (T.I.), qualifier ions (Q.I) and retention times (R.T) listed in Table 3.4.

Table 3.2. Operating Conditions of Thermal Desorption System

Purge Time	1 minute
Inject Time	1 minute
Tube Desoption Temperature	250 °C
Tube Desorption Time	3 minutes
Trap Low Temperature	- 10 °C
Trap High Temperature	300 °C
Trap Hold Time	3 minutes

Table 3.3. Gas Chromatography/Mass Spectrometry (GC/MS) Operating Parameters

Column	HP-VOC 60m. 320μm. 1.80μm
Gos Chromotography Model	Agilent Technologies Inc. 6890N Gas
Gas Chromatography Model	Chromatograph
Carrier Gas Type	Hellium 1.1ml/min (constant flow)
Initial Oven Temperature	40 °C
Override Solvent Delay Time	8 minutes
Initial Time	10 minutes
Oven Ramp Rate	8 minutes
Final Temperature	250 °C
Final Time	5 minutes
Initial GC inlet Temperature	240 °C
Pressure	0.377 bar
Purge Flow	50.0 ml/min
Run Time	41.25 minutes

When tube analysis is completed, tube was removed from the thermal desorber and, using clean gloves, the Teflon® caps with Swagelok fittings and PTFE were recapped and the tubes were re-store in a cool environment (<4 °C).

Table. 3.4. Chemical Properties of Target VOC Compounds

Analyte	M.W	Boiling Point/	R.T ^a	T.I ^b	Q.I ^c
	(g/mol)	Range (°C)			
2-propanol	60.1	82	7.849	45	27
methylene chloride	84.93	39.6	9.852	49	84
1-propanol	60.1	97	10.994	31	29
hexane	86.18	68-69	12.731	57	41
2-butanone	72.11	79.64	13.262	43	72
ethyl acetate	88.11	77.1	14.074	43	29
chloroform	119.38	61.2	14.452	83	85
1,1,1 trichloroethane	133.40	74	15.679	97	99
1,2 dichloroethane	98.96	84	16.114	62	64
n-butanol	74.12	118	16.365	56	41
carbon tetrachloride	153.82	76.72	16.430	117	119
benzene	78.11	80.1	16.495	78	77
1so-octane	114.23	99	17.100	57	41
n-heptane	100.21	98-99	17.583	43	41
trichloroethylene	131.39	87.2	18.127	130	132
1,2 dichloropropane	112.99	95-96	18.225	63	41
bromodichloromethane	163.83	90	18.595	83	85
4-methyl-2-pentanone	100.16	117-118	19.649	43	58
toluene	92	111	20.785	91	92
n-octane	114.23	125-126	21.349	43	41
n-butyl acetate	116.16	126-127	22.041	43	56
dibromochloromethane	208.28	119-120	22.041	129	127
tetrachloroethene	165.83	121.2	22.262	166	164
ethylbenzene	106.17	136	23.869	91	106
p-m xylene	106.2	138-139	24.089	91	106
n-nonane	128.26	150-151	24.361	45	57
styrene	104	145	24.797	104	78
o-xylene	106.2	144	24.858	91	106
(1s)-(-)α-pinene	136.23	155	25.864	93	91
3-ethyltolune	120.19	158-159	26.585	105	120/91
4-ethyltolune	120.19	162	26.642	105	120/106
1,3,5 trimethylbenzene	120.19	163-165	26.757	105	120
n-decane	142.28	174	26.928	57	43
(-)-β-pinene	136.23	165-167	27.143	93	41
2-ethyltolune	120.19	164-165	27.143	105	120
1,2,4 trimethylbenzene	120.19	169-171	27.473	105	120
(r)-(+) limonene	136.24	176	28.216	68	93
1,4 dichlorobenzene	147	174.12	28.216	146	148
1,2,3 trimethylbenzene	120.19	175-176	28.298	105	120
n-undecane	156.31	196	29.205	57	43
nonanal	142.24	93	29.668	41	57
1,2,4,5 tetramethylbenzene	134.218	192-197	30.347	119	134

Table. 3.4. (cont.)

Analyte	M.W (g/mol)	Boiling Point/ Range (°C)	R.T ^a	T.I ^b	Q.I ^c
n-dodecane	170.33	214-218	31.289	57	43
decanal	156.2	207-209	31.777	41	57
n-tridecane	184.36	232-236	33.216	57	43
n-tetradecane	198.388	252-254	35.017	57	43
n-pentadecane	212.41	268-270	36.716	57	43
n-hexadecane	226.44	271-291	38.508	57	43

^aRT: Retention Time, ^bTI: Target Ion, ^cQI: Qualifier Ion

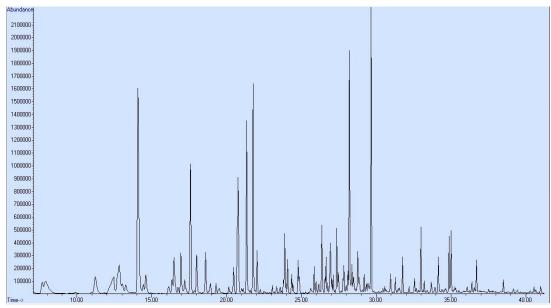


Figure 3.3. An Example Gas Chromatogram

3.2.2. Aldehyde Analysis

Aldehyde analysis was performed in two stages. The first stage is *Extraction* of sample collected DNPH silica gel sorbent tubes using 99.9% purity acetonitrile (Merck 1.00030). Five ml stainless steel gas tight syringes (Agilent Gas Tight Luer Lock Valve 5183-4552 and 4451) was used to flow acetonitrile through the cartridge. Sample eluted from the cartridge was collected in to 40-ml vials (National Scientific, Screw Caps with PTFE Septa, B7999-6A). Then, 3 ml of effluent transferred to 4 ml vials (National Scientific, Screw Caps with PTFE Septa, C4015-1). All glassware were rinsed with acetonitrile for cleaning after then were heated in 60 °C vacuum oven at least 60

minutes. Extraction was performed based on the guidance of EPA Method IP-6A. The steps of extraction described below respectively.

(1) DNPH silica gel tubes were removed from refrigerator with clean gloves then, aluminum foil was unwrapped and sealed caps were removed. (2) The DNPH silica gel sorbent tube was attached to a stand in vertically in the reverse direction of the arrow on the tube. (3) The aldehyde derivative was eluted from the tube with 6 ml of acetonitrile. (4) Effluent from the sampler was collected in a 40 ml vial. (5) 3ml of effluent was transferred to a 4 ml vial and capped with a chemically inert cap. (6) The vial was labeled for identification and was wrapped with aluminum foil to protect from the light. (7) The 4 ml vial was placed in the refrigerator and was kept until HPLC analysis. (8)Sample eluates are stable at 4 °C for up to one month.

The second stage is *HPLC analysis*. Calibration of the HPLC system (Agilent 1100 series, Japan) was performed as described in Section 3.2.3.1. Analysis of sample eluates were performed in one week with an HPLC equipped with a Ultraviolet Visible (UV) absorption detector operated at 360 nm for detemination of target aldehydes. The operating parameters are summarized in Table 3.5. Before analysis, the sample was diluted with water. One ml sample was tranferred to another clean 4 ml vial and 1 ml water was added to the vial. From this diluted sample 20 µl was injected to the HPLC instrument. All target aldehydes in the sample were identified and quantified by comparison of their retention times (Table 3.6) and peak heights or peak areas with those of standard DNPH derivatives. The linear gradient program varied the mobile phase composition periodically. The following gradient program was found to be adequate to achieve this goal: after 20 µl sample injection, linear gradient was stable at 70% acetonitrile / 30% water in 10 minutes, linear gradient from 70-100% acetonitrile / 30-0% water in 20 minutes, reverse gradient from 100-70% acetonitrile / 0-30% water in 2 minutes then hold at 70% acetonitrile for 3 minutes to be ready for next injection.

Table 3.5. Operating Parameters of High Performance Liquid Chromatograph (HPLC)

Column	GL Sciences, Inertsil ODS 3 (4.6 mm x 250 mm x 5 μm)
Mobile Phase	70% Acetonitrile (A)/ 30% Water (W), Gradient Program
Gradient	0 min (70% A), 10 min (70% A), 30 min (100% A), 32 min(70% A), 35 min (70% A)
Detector	UV/VIS, operating at 360nm
Flow Rate	1 ml/min
Operating Temperature	25 °C
Sample Injection Volume	20 μ1

Table 3.6. Chemical Propperties of Target Aldehyde Compounds

M.W(g/mol)	R.T ^a
30.03	5.35
44.05	6.47
56.06	7.65
58.08	8.05
58.08	8.50
70.09	9.90
72.11	11.10
106.12	12.15
86.13	14.35
86.13	14.85
120.15	15.00
120.15	16.00
100.15	18.45
134.18	18.85
	30.03 44.05 56.06 58.08 58.08 70.09 72.11 106.12 86.13 86.13 120.15 120.15 100.15

^aRT: Retention Time

3.2.3. Quality Assurance/Quality Control

3.2.3.1. Calibration Standards

Japanese Indoor Air Standard Mix 100 μg/mL in methanol: water (19:1), 52 Chemicals, (Supelco Catalog No: 4M7537-U) was used for GC-MS calibration. For the standard preparation process a solvent which is pure and contain less than 10% of minumum analyte levels and more volatile than target compounds must be used thus during standard preparation process solvent is purged and eliminated from the Tenax

TA tube. Methanol was the most suitable solvent that has the essential criteria therefore it was used as solvent to prapare the calibration standards of VOC mixtures in this study. Eight levels of calibration standards (10, 20,50, 100, 200, 500, 1000, 2000 μ g l⁻¹) were used to prepare the calibration curves. For all compounds the linear fit was satisfactory (R²>0.99).

Aldehydes calibration was performed by direct injection of standard mixtures with known amounts of solid hydrazones dissolved in acetonitrile. The calibration curve was made using a certified stock solution containing a mixture of 15 hydrazones from 15 μ g/mL (Supelco catalog number 4M7285-U). The certified stock solution includes the hydrazones derivated of the following compounds: formaldehyde, acetaldehyde, acrolein, acetone, propionaldehyde, crotonaldehyde, butyraldehyde, benzaldehyde, isovaleraldehyde, valeraldehyde, otolualdehyde, m-tolualdehyde, p-tolualdehyde, 2,5-dimetilbenzaldehyde, and hexaldehyde. The target compounds were identified by comparison of their chromatographic retention times with those of the derivatized standards. Standards were prepared at least at four concentration levels between 7 and 5000 μ g/l and for each target compound a calibration curve was generated by linear regression of the concentration and chromatographic response data. The calibration curves were good linear fits (R^2 >0.99).

3.2.3.2. Blanks

Based on the U.S. EPA guidance, laboratory or field blanks must be included in the study. Based on this criteria, field blanks were included in this study. Field blanks are conditioned at the same time as the samples. The transportation and storage of field blank tubes were carried out with the sample tubes as described in section 3.1.5. At sampling location, with clean gloves field blank tubes were removed from storage container, the sealed caps of DNPH silica gel sorbent tube and Swagelok® type unions and combined PTFE ferrules of Tenax TA tubes were taken off. The field blank tubes were then attached to the sampling lines with non outgassing flexible tubing. Immediately after this, the tubes were recapped. The field blank tubes were placed back in the storage container and transported back to the laboratory with samples. The point to be considered, air was not pumped through field blank tubes. The number of field blanks were determined as 10% of the total sample size (21 sampling tubes total each

for Tenax TA and DNPH). Therefore, three Tenax TA field blanks and three DNPH silica gel sorbent tube field blanks were collected. The field blanks were put through the processes for sample analysis. Field blank aldehyde concentrations are presented in Table 3.7. Four of the twelve aldehydes were detected in field blank samples. The rest of the aldehydes were at below their respective the detection limits. In Table 3.8 Field Blank VOC concentrations are shown, twenty seven VOCs were detected in field blank samples. The rest of the twenty seven VOCS were at below their respective the detection limits.

Table 3.7. Aldehyde Amounts in Field Blanks

Target Aldehyde	Field Blank Amounts (μg/m ³)
formaldehyde	0.68
acetaldehyde	0.17
propionaldehyde	BDL
crotonaldehyde	BDL
butyraldehyde	0.17
benzaldehyde	BDL
ısovaleraldehyde	BDL
valeraldehyde	BDL
o-tolualdehyde	BDL
<i>m</i> -p-tolualdehyde	0.03
hexaldehyde	BDL
2,5-dimethylbenzaldehyde	BDL

BDL: Below Detection Limit

Limit of Quantification (LOQ) which is lowest actual amount of analyte that can be reliably quantified for all target VOC compounds and target aldehyde compounds were determined. The LOQ values are given in Table 3.9 and Table 3.10 for VOCs and aldehydes respectively.

Table 3.8. VOC Amounts in Field Blanks

Target VOC	Field Blank Amounts (μg/m³)
1,2,4-trimethylbenzene	0.01
ethyl acetate	0.85
4methyl2pentanone	BDL
toluene	0.27
n-octane	0.11
dibromochloromethane	BDL

Table. 3.8. (cont.)

Target VOC	Field Plants Amounts (a/m3)
Target VOC	Field Blank Amounts (µg/m³)
n-butyl acetate	<0.01 BDL
tetrachloroethene	
ethylbenzene	0.01
p-m-xylene	0.04
n-nonane	0.02
styrene	0.01
o-xylene ·	BDL
a-pinene	0.01
3-ethyltoluene	<0.01
4-ethyltoluene	BDL
1,3,5-trimethylbenzene	BDL
n-decane	BDL
b-pinene	BDL
2-ethyltoluene	0.03
1,4-dichlorobenzene	0.01
limonene	BDL
1,2,3-trimethylbenzene	BDL
n-undecane	1.01
nonanal	0.64
1,2,4,5-tetramethylbenzene	BDL
n-dodecane	< 0.01
decanal	0.42
n-tridecane	< 0.01
n-tetradecane	< 0.01
n-pentadecane	BDL
n-hexadecane	BDL
hexane	0.03
2-butanone	BDL
chloroform	0.01
1,1,1-trichloroethane	N.D.
1,2-dichloroethane	< 0.01
n-butanol	BDL
carbontetrachloride	< 0.01
benzene	0.63
iso-octane	0.09
n-heptane	BDL
tricloroethylene	< 0.01
1,2-dichloropropane	BDL
bromodichloromethane	BDL
2-propanol	BDL
methylene chloride	0.30
1-propanol	BDL
	ı

BDL: Below Detection Limit

N.D: Not Detected

Table 3.9. Limits of Quantification (LOQ) for Target VOC Compounds

Target Compounds	LOQ $(\mu g/m^3)$	Target Compounds	LOQ $(\mu g/m^3)$
1,2,4-trimethylbenzene	0.0025	1,2,4,5-tetramethylbenzene	0.0004
ethyl acetate	0.006	n-dodecane	0.0038
4 methyl 2 pentanone	0.0051	decanal	0.1799
toluene	0.0086	n-tridecane	0.0064
n-octane	0.0239	n-tetradecane	0.0077
dibromochloromethane	0.001	n-pentadecane	0.0180
n-butyl acetate	0.003	n-hexadecane	0.0148
tetrachloroethene	0.001	hexane	0.0039
ethylbenzene	0.0071	2-butanone	0.0247
p-m-xylene	0.0053	chloroform	0.012
n-nonane	0.0378	1,1,1-trichloroethane	0.0114
styrene	0.0125	1,2-dichloroethane	0.0078
o-xylene	0.0005	n-butanol	0.0140
a-pinene	0.001	carbontetrachloride	0.0066
3-ethyltoluene	0.0013	benzene	0.5719
4-ethyltoluene	0.0006	ıso-octane	0.0222
1,3,5-trimethylbenzene	0.0006	n-heptane	0.0283
n-decane	0.0067	tricloroethylene	0.0081
b-pinene	0.004	1,2-dichloropropane	0.0086
2-ethyltoluene	0.0008	bromodichloromethane	0.001
1,4-dichlorobenzene	0.0086	2-propanol	0.0196
limonene	0.0010	methylene chloride	0.0246
1,2,3-trimethylbenzene	0.0006	1-propanol	0.0590
n-undecane	0.0042		
nonanal	0.1545		

Table 3.10. Limits of Quantification (LOQ) for Target Aldehyde Compounds

Target Compund	LOQ (µg/m3)
formaldehyde	0.0225
acetaldehyde	0.0197
acrolein	0.0147
acetone	0.0247
propionaldehyde	0.0361
crotonaldehyde	0.0315
butryaldehyde	0.0238
benzaldehyde	0.0298
ısovaleraldehyde	0.0365
valeraldehyde	0.0197
o-tolualdehyde	0.0431
m/p-tolualdehyde	0.0185
hexaldehyde	0.0271
2,5-dimethylbenzaldehyde	0.0183

3.2.3.3. Performance Criteria for The Pump

Calibration of the pumps were performed before each sample. At the end of the sampling, the flow rate of the pumps were checked with the flow calibrator. In EPA Method TO-17 and EPA Method IP-6A is stated that diffrence between sampling flow rate measured at the end of sample collection that measured at the beginning of sample collection must be less than 10% for the sample to be considered valid. In this study, difference between the beginning and the end flow rate of the pumps were less than 10%. The differences were as $\pm 4\%$ and $\pm 5\%$ for VOCs and aldehydes, respectively.

3.2.3.4. Duplicate Precision

Duplicate precision measurement is the absolute value of the relative difference between two identical parallel samples (same flow rate over the same time period from with a common inlet to the sample volume). In this study, duplicate precision measurement was performed for VOCs and aldehydes with two identical Tenax TA and DNPH tubes were used in parallel during sampling. In the analytical precision of duplicate pairs is expressed as a percentage as follows in Equation 3.1:

Analytical Precision:
$$=\frac{[X1-X2]}{X}*100$$
 (3.1)

where:

 $X_1 = A$ measurement value taken from one of the two tubes using in sampling.

 $X_2 = A$ measurement value taken from the second of two tubes using in sampling.

 $X = Average of X_1 and X_2.$

It is recommended in EPA Method TO-17 duplicate (analytical) precision within 25% for distributed volume pairs of tubes. Analytical precision of duplicate pairs of 39 target VOCs were found within 25% but analytical precision of duplicate pairs of 8 target VOCs (p-m xylene, styrene, 1,2,4,5 tetramethylbenzene, n-dodecane, hexane, 1,1,1 trichloroethane, 2-propanol, and 1-propanol) were found higher than 25% in this study. The concentrations of these 8 VOCs were found in very low levels, therefore the

uncertainty arising from this result should not be significant. Agreement between the duplicate pairs of aldehydes were found 8.4% and 20.7% for formaldehyde and acetaldehyde, respectively. Rest of target aldehydes were found in low concentration levels ($<1~\mu g/m^3$), as a result, for which the analytical precision were found to be higher than 25%. The arising uncertainty should not be significant because the concentrations were low.

CHAPTER 4

RESULTS

In this section the results are presented in two main sections. The first section includes sampling results of target VOCs, aldehydes, and PM_{2.5}, and measurement results of PM₁₀, TVOC, CO₂, CO, temperature, and relative humidity in the restaurant kitchen before frying, during frying, and after frying during regular operation (Campaign-1) to characterize the cook's exposure concentrations. In the second section, results of Campaign-2 are presented. The same sampling and measurements were performed again in Campaign-2 this time under controlled conditions: only French cut potatoes were fried at daily varying amounts (1st day, 1250 g; 2nd day, 2500 g; and 3rd day, 3750 g). Discussion of the results are presented in the next chapter.

4.1 Campaign-1, Sampling

Samples were collected for three days in the restaurant kitchen. The campaign was started on 31st of October and ended on 15th of November. Fried products were mostly French fries but also nuggets, schnitzels, hamburger patties, and chicken burger patties were fried in the deep fryer.

4.1.1. VOC Concentrations

In the first campaign of the study, 47 target VOC compounds were detected in all sampling periods in the restaurant kitchen. Only 2-butanone was not detected in some samples (non-detects are denoted with Below Detection Limit, BDL). Summary statistics for before frying, during frying, and after frying sampling period VOC concentrations are given in Table 4.1, Table 4.2, Table 4.3, respectively.

Concentrations of 22 VOCs were increased in during frying sampling period, which were ethyl acetate, toluene, n-octane, n-butyl acetate, tetrachloroethene, n-

nonane, n-decane, 1,4-dichlorobenzene, n-undecane, nonanal, decanal, n-tridecane, n-pentadecane, n-hexadecane, hexane, 1,1,1-trichloroethane, 1,2-dichloroethane, n-butanol, n-heptane, 1,2-dichloropropane, 1-propanol. Total volatile organic compound concentration was calculated by the sum of the target VOC compounds (Σ TVOC). The mean of Σ TVOC concentration in the periods of before, during, and after frying were 139 μ g/m³, 208 μ g/m³, and 144 μ g/m³, respectively. The median Σ TVOC concentrations were similar. It can be concluded that the Σ TVOC concentration increased about 50% due to the frying activity. In other words, the cooks may be exposed to VOCs 1.5 times the background concentrations when they fry using the vegetable margarine.

Table 4.1. Summary Statistics of Before Frying Period VOC Concentrations $(\mu g/m^3)$ in the Restaurant Kitchen (n=3)

Target Compounds	Mean	Median	SD	Min	Max
1,2,4-trimethylbenzene	0.42	0.43	0.08	0.33	0.49
ethyl acetate	20.0	17.8	10.3	11.0	31.2
4 methyl 2 pentanone	0.15	0.07	0.13	0.07	0.29
toluene	4.18	4.02	0.56	3.72	4.80
n-octane	5.24	4.88	1.65	3.80	7.03
dibromochloromethane	0.02	0.02	0.01	0.02	0.03
n-butyl acetate	2.33	2.65	0.90	1.31	3.04
tetrachloroethene	0.20	0.22	0.08	0.12	0.26
ethylbenzene	0.43	0.40	0.05	0.39	0.49
p-m-xylene	0.59	0.61	0.06	0.52	0.62
n-nonane	0.67	0.63	0.23	0.46	0.91
Styrene	0.42	0.33	0.19	0.30	0.64
o-xylene	0.45	0.49	0.09	0.35	0.52
α-pinene	2.27	1.58	1.33	1.43	3.81
3-ethyltoluene	0.29	0.30	0.04	0.25	0.32
4-ethyltoluene	0.13	0.14	0.02	0.11	0.15
1,3,5-trimethylbenzene	0.08	0.09	0.01	0.07	0.09
n-decane	1.24	0.93	0.64	0.81	1.97
β-pinene	1.20	1.40	0.41	0.72	1.46
2-ethyltoluene	0.13	0.13	0.02	0.11	0.15
1,4-dichlorobenzene	2.26	1.53	1.74	0.99	4.25
limonene	37.4	20.7	30.4	19.1	72.5
1,2,3-trimethylbenzene	0.11	0.11	0.00	0.10	0.11
n-undecane	0.09	0.09	0.01	0.08	0.10
nonanal	13.7	13.7	1.59	12.1	15.3
1,2,4,5-tetramethylbenzene	0.01	0.01	0.00	0.01	0.01
n-dodecane	0.34	0.28	0.15	0.22	0.51
decanal	2.31	2.34	0.23	2.07	2.52
n-tridecane	0.38	0.27	0.23	0.22	0.64
n-tetradecane	2.13	1.65	1.42	1.01	3.73

Table 4.1. (cont.)

Target Compounds	Mean	Median	SD	Min	Max
n-pentadecane	0.74	0.80	0.14	0.58	0.85
n-hexadecane	0.66	0.60	0.20	0.50	0.89
hexane	0.65	0.53	0.57	0.16	1.27
2-butanone	BDL	BDL	< 0.01	< 0.01	< 0.01
chloroform	0.97	0.50	1.07	0.21	2.19
1,1,1-trichloroethane	0.01	0.01	0.01	< 0.01	0.02
1,2-dichloroethane	0.45	0.41	0.36	0.11	0.82
n-butanol	0.83	0.84	0.05	0.77	0.86
carbontetrachloride	0.79	0.84	0.15	0.62	0.90
benzene	5.73	4.87	4.03	2.20	10.11
iso-octane	2.88	1.73	3.13	0.49	6.42
n-heptane	23.6	24.0	9.35	14.1	32.8
tricloroethylene	0.15	0.12	0.13	0.03	0.29
1,2-dichloropropane	< 0.01	< 0.01	< 0.01	< 0.01	0.01
bromodichloromethane	0.05	0.03	0.06	0.01	0.12
2-propanol	0.55	0.24	0.65	0.11	1.29
methylene chloride	1.92	1.45	2.03	0.17	4.14
1-propanol	0.12	0.12	0.02	0.10	0.13
ΣΤΥΟС	139	134	13.0	129	154

BDL: Below Detection Limit

Table 4.2. Summary Statistics of During Frying Period VOC Concentrations ($\mu g/m^3$) in the Restaurant Kitchen (n=3)

Target Compounds	Mean	Median	SD	Min	Max
1,2,4-trimethylbenzene	0.41	0.41	0.02	0.39	0.43
ethyl acetate	24.8	29.6	13.9	9.20	35.7
4 methyl 2 pentanone	0.12	0.12	0.08	0.06	0.18
toluene	4.37	4.16	0.41	4.10	4.85
n-octane	16.6	15.9	11.4	5.6	28.4
dibromochloromethane	0.02	0.02	< 0.01	0.02	0.02
n-butyl acetate	2.80	3.01	0.54	2.18	3.21
tetrachloroethene	0.22	0.22	0.08	0.14	0.29
ethylbenzene	0.38	0.35	0.08	0.31	0.47
p-m-xylene	0.53	0.50	0.06	0.49	0.59
n-nonane	1.07	1.06	0.53	0.55	1.60
styrene	0.30	0.31	0.03	0.26	0.32
o-xylene	0.41	0.41	0.07	0.34	0.47
α-pinene	1.64	1.30	0.62	1.27	2.36
3-ethyltoluene	0.28	0.28	0.00	0.28	0.29
4-ethyltoluene	0.13	0.13	0.01	0.12	0.13
1,3,5-trimethylbenzene	0.08	0.08	< 0.01	0.08	0.08
n-decane	1.91	1.22	1.58	0.79	3.72

Table 4.2. (cont.)

Target Compounds	Mean	Median	SD	Min	Max
β-pinene	0.81	0.70	0.38	0.49	1.23
2-ethyltoluene	0.12	0.11	0.01	0.11	0.13
1,4-dichlorobenzene	2.66	2.73	1.70	0.93	4.32
limonene	21.9	14.5	18.6	8.33	43.2
1,2,3-trimethylbenzene	0.11	0.11	0.01	0.10	0.11
n-undecane	0.40	0.40	0.22	0.24	0.55
nonanal	23.4	23.6	6.48	16.9	29.8
1,2,4,5-tetramethylbenzene	0.01	0.01	< 0.01	0.01	0.02
n-dodecane	0.49	0.36	0.34	0.23	0.87
decanal	3.12	3.03	0.71	2.46	3.87
n-tridecane	0.41	0.28	0.27	0.23	0.72
n-tetradecane	1.93	1.80	1.08	0.93	3.07
n-pentadecane	0.89	0.96	0.18	0.68	1.02
n-hexadecane	0.69	0.57	0.20	0.57	0.92
hexane	0.90	0.67	0.43	0.63	1.39
2-butanone	0.01	0.01	< 0.01	0.01	0.01
chloroform	0.75	0.38	0.81	0.19	1.68
1,1,1-trichloroethane	1.54	0.01	2.65	0.00	4.60
1,2-dichloroethane	0.49	0.45	0.18	0.34	0.69
n-butanol	0.88	0.78	0.18	0.77	1.08
carbontetrachloride	0.71	0.63	0.18	0.60	0.92
benzene	3.88	4.22	0.93	2.82	4.59
iso-octane	0.10	0.10	BDL	0.10	0.10
n-heptane	83.0	79.9	54.2	30.4	139
tricloroethylene	0.12	0.11	0.11	0.02	0.23
1,2-dichloropropane	0.01	0.01	< 0.01	< 0.01	0.01
bromodichloromethane	0.03	0.03	0.02	0.01	0.05
2-propanol	0.13	0.12	0.04	0.09	0.17
methylene chloride	1.83	0.91	2.23	0.22	4.38
1-propanol	0.15	0.16	0.03	0.11	0.17
ΣΤVΟC	207	196	83.1	131	296

Table 4.3. Summary Statistics of After Frying Period VOC Concentrations ($\mu g/m^3$) in the Restaurant Kitchen (n=3)

Target Compounds	Mean	Median	SD	Min	Max
1,2,4-trimethylbenzene	0.41	0.33	0.19	0.28	0.63
ethyl acetate	23.5	20.2	13.9	11.5	38.8
4 methyl 2 pentanone	0.33	0.15	0.33	0.13	0.72
toluene	2.92	2.98	0.61	2.29	3.51
n-octane	5.97	5.63	2.14	4.01	8.26
dibromochloromethane	0.06	0.06	0.01	0.05	0.07
n-butyl acetate	3.03	3.36	1.03	1.88	3.85

Table 4.3. (cont.)

Target Compounds tetrachloroethene ethylbenzene p-m-xylene	Mean 0.09 0.31	Median 0.09	SD 0.03	Min 0.07	Max
ethylbenzene		0.09	0.03	0.07	0.10
	0.31			0.07	0.13
n-m-x vlene		0.27	0.13	0.21	0.46
p iii Ajiciic	0.43	0.39	0.16	0.31	0.61
n-nonane	1.02	1.06	0.38	0.62	1.39
styrene	0.17	0.15	0.04	0.14	0.22
o-xylene	0.36	0.32	0.18	0.21	0.56
α-pinene	1.17	1.13	0.17	1.02	1.35
3-ethyltoluene	0.29	0.22	0.15	0.19	0.47
4-ethyltoluene	0.13	0.10	0.07	0.08	0.21
1,3,5-trimethylbenzene	0.10	0.07	0.06	0.06	0.16
n-decane	1.21	1.28	0.47	0.71	1.64
β-pinene	0.38	0.36	0.13	0.26	0.51
2-ethyltoluene	0.12	0.09	0.06	0.07	0.19
1,4-dichlorobenzene	13.1	13.2	11.4	1.66	24.4
limonene	16.4	16.2	12.7	3.73	29.2
1,2,3-trimethylbenzene	0.11	0.09	0.05	0.08	0.16
n-undecane	0.12	0.11	0.03	0.09	0.15
nonanal	19.9	14.1	11.1	12.8	32.6
1,2,4,5-tetramethylbenzene	0.01	0.01	0.01	< 0.01	0.02
n-dodecane	0.36	0.27	0.19	0.24	0.58
decanal	2.92	2.41	1.64	1.60	4.75
n-tridecane	0.33	0.33	0.12	0.21	0.44
n-tetradecane	1.39	1.22	0.71	0.78	2.18
n-pentadecane	0.64	0.60	0.10	0.56	0.75
n-hexadecane	0.60	0.56	0.11	0.51	0.72
hexane	0.46	0.42	0.24	0.24	0.71
2-butanone	BDL	BDL	BDL	BDL	BDL
chloroform	7.72	1.77	10.81	1.20	20.21
1,1,1-trichloroethane	0.01	0.01	0.01	< 0.01	0.02
1,2-dichloroethane	0.84	0.32	1.09	0.11	2.09
n-butanol	0.76	0.75	0.06	0.71	0.82
carbontetrachloride	0.90	0.96	0.29	0.59	1.17
benzene	2.22	1.89	0.78	1.65	3.11
iso-octane	1.12	1.12	0.14	1.02	1.22
n-heptane	28.7	26.4	7.93	22.2	37.6
tricloroethylene	0.05	0.06	0.01	0.04	0.06
1,2-dichloropropane	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01
bromodichloromethane	0.31	0.26	0.27	0.06	0.61
2-propanol	0.18	0.12	0.11	0.11	0.31
methylene chloride	3.41	1.73	4.31	0.19	8.32
1-propanol	0.22	0.22	0.12	0.11	0.35
ΣΤΥΟΣ	144	135	24.7	126	172

BDL: Below Detection Limit

In order to have a closer look on the change in the concentrations before, during, and after frying periods, a graph of eight VOCs with the highest increase in the mean indoor concentrations is drawn (Figure 4.1). Before frying concentrations were 23.6, 20.0, 13.7, 5.24, 4.18, 2.31, 1.24, 0.65 $\mu g/m^3$, during frying concentrations were 83.0, 24.9, 23.4, 16.6, 4.37, 3.12, 1.91, 0.90 $\mu g/m^3$, and after frying concentrations were 28.7, 23.5, 19.9, 5.97, 2.92, 2.92, 1.21, 0.46 $\mu g/m^3$ for n-heptane, ethyl acetate, nonanal, n-octane, toluene, decanal, n-decane, and hexane respectively. According to these results, the highest increase in during frying sampling period was from 23.6 $\mu g/m^3$ to 83.0 $\mu g/m^3$ (3.5 folds) in n-heptane concentrations. It was followed by 3.2, 1.7, and 1.25 folds by n-octane, nonanal, and ethyl acetate, respectively. The concentrations were decreased in the after frying period, but did not drop back to the background levels in 1.5 hours.

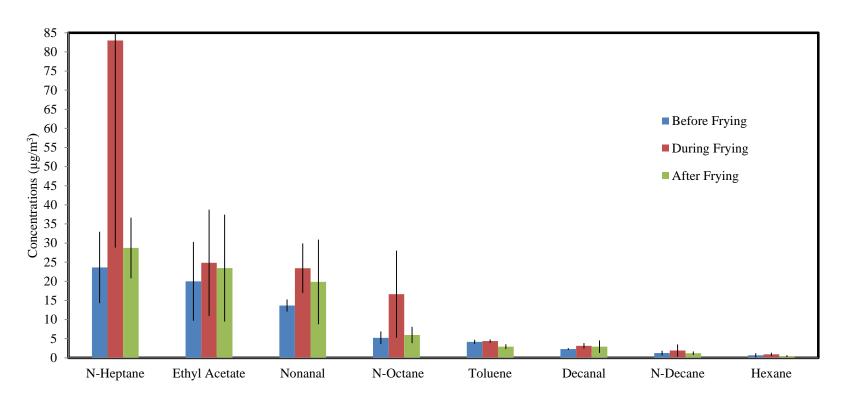


Figure 4.1 VOCs Concentrations with the Highest Increase in During Frying Period in The Restaurant Kitchen

4.1.2. Aldehyde Concentrations

Fifteen aldehydes (formaldehyde, acetaldehyde, acrolein, acetone, propionaldehyde, crotonaldehyde, butryaldehyde, benzaldehyde, isovaleraldehyde, valeraldehyde, o-tolualdehyde, m/p-tolualdehyde, hexaldehyde, 2.5dimethylbenzaldehyde) were targeted in this study. Fourteen of those were detected in all periods of sampling except for Acrolein in the restaurant kitchen. Acrolein is a highly instable aldehyde and it is usually converted to a more stable compound for analysis so it is difficult to analyze with direct methods (Fullana et al. 2004). Acrolein could not be determined in all of our samples because the method that we used for aldehyde sampling is considered to be inappropriate to investigate acrolein. Therefore, acrolein concentrations are not reported. In addition to this acetone concentrations were not included because the capacity of the sorbent tube was considered to be not sufficient.

Concentrations of eight aldehydes increased in during frying sampling period, which were acetaldehyde, formaldehyde, crotonaldehyde, butryaldehyde, valeraldehyde hexaldehyde and 2.5-dimethylbenzaldehyde. Also, total aldehydes were determined by summing the detected target aldehyde compounds. The mean concentrations of total aldehydes were calculated as $11.9 \,\mu\text{g/m}^3$ for before frying period, $19.4 \,\mu\text{g/m}^3$ for during frying period, and $24.5 \,\mu\text{g/m}^3$ for after frying period. The median total aldehyde concentrations were $12.1 \,\mu\text{g/m}^3$, $15.5 \,\mu\text{g/m}^3$, $16.8 \,\mu\text{g/m}^3$ for before frying, during frying, and after frying periods respectively. It can be concluded that the total aldehyde concentration was increased about 50 % due to the frying activity. The results and summary statistics of before frying, during frying and after frying aldehyde concentrations are given in Table 4.4, Table 4.5 and Table 4.6 respectively. Mean and median aldehyde concentrations in during frying period were close to each other. The highest difference between mean and median values was found for acetaldehyde as $2 \,\mu\text{g/m}^3$.

Aldehydes that had the highest increase in concentration from before to during frying periods were acetaldehyde, formaldehyde and hexaldehyde, respectively. Their concentrations in the period of before frying were determined as 6.64, 2.66, and 0.78 $\mu g/m^3$, in the period of during frying the concentrations were 13.1, 2.95, and 1.29 $\mu g/m^3$, and in after frying sampling mean concentrations were 17.3, 4.18 and 1.05

 μ g/m³ respectively. Figure 4.2 shows the change in the three highest mean concentration aldehydes before, during, and after frying periods.

Table 4.4. Summary Statistics of Before Frying Period Aldehyde Concentrations ($\mu g/m^3$) in the Restaurant Kitchen (n=3)

Target Compound	Mean	Median	SD	Min	Max
formaldehyde	2.66	2.76	0.27	2.35	2.86
acetaldehyde	6.64	6.04	2.03	4.98	8.90
propionaldehyde	0.42	0.24	0.31	0.23	0.78
crotonaldehyde	0.03	0.04	0.02	0.00	0.04
butryaldehyde	0.24	0.29	0.11	0.11	0.32
benzaldehyde	0.28	0.28	0.04	0.25	0.32
isovaleraldehyde	0.19	0.15	0.07	0.15	0.27
valeraldehyde	0.28	0.41	0.24	0.00	0.42
o-tolualdehyde	0.12	< 0.01	0.22	0.00	0.37
m/p-tolualdehyde	0.12	0.10	0.13	0.00	0.26
hexaldehyde	0.78	0.86	0.25	0.50	0.97
2,5-dimethylbenzaldehyde	0.11	0.08	0.08	0.05	0.19
total aldehydes	11.9	12.1	2.83	8.91	14.5

Table 4.5. Summary Statistics of During Frying Period Aldehyde Concentrations ($\mu g/m^3$) in the Restaurant Kitchen (n=3)

Target Compound	Mean	Median	SD	Min	Max
formaldehyde	2.95	2.93	1.35	1.61	4.32
acetaldehyde	13.1	11.1	11.0	3.34	25.0
propionaldehyde	0.39	0.47	0.14	0.23	0.47
Crotonaldehyde	0.04	< 0.01	0.07	< 0.01	0.12
butryaldehyde	0.25	0.20	0.11	0.18	0.38
benzaldehyde	0.27	0.26	0.06	0.21	0.34
isovaleraldehyde	0.14	0.19	0.12	< 0.01	0.23
valeraldehyde	0.46	0.43	0.06	0.41	0.52
o-tolualdehyde	0.12	0.12	0.13	< 0.01	0.25
m/p-tolualdehyde	0.11	0.13	0.04	0.07	0.14
hexaldehyde	1.29	1.23	0.15	1.18	1.46
2,5-dimethylbenzaldehyde	0.22	0.21	0.01	0.21	0.23
total aldehydes	19.4	15.5	10.3	11.6	31.0

Table 4.6. Summary Statistics of After Frying Period Aldehyde Concentrations ($\mu g/m^3$) in the Restaurant Kitchen (n=3)

Target Compound	Mean	Median	SD	Min	Max
formaldehyde	4.18	2.55	3.44	1.86	8.14
acetaldehyde	17.3	4.06	24.8	2.01	45.9
propionaldehyde	0.30	0.31	0.10	0.19	0.38
crotonaldehyde	0.04	< 0.01	0.07	< 0.01	0.12
butryaldehyde	0.27	0.16	0.26	0.08	0.57
benzaldehyde	0.30	0.32	0.07	0.23	0.36
isovaleraldehyde	0.19	0.22	0.18	0.00	0.36
valeraldehyde	0.49	0.36	0.36	0.21	0.89
o-tolualdehyde	0.05	< 0.01	0.08	< 0.01	0.14
m/p-tolualdehyde	0.17	0.09	0.15	0.07	0.34
hexaldehyde	1.05	0.87	0.37	0.81	1.47
2,5-dimethylbenzaldehyde	0.13	0.11	0.04	0.10	0.17
total aldehydes	24.5	16.8	23.2	6.13	50.6

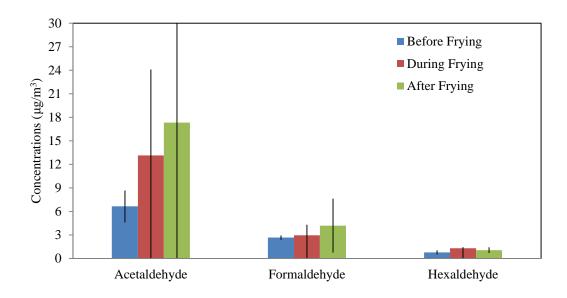


Figure 4.2. Aldehyde Concentrations with Highest Increase in During Frying Period in the Restaurant Kitchen

4.1.3. PM_{2.5} Concentrations

 $PM_{2.5}$ concentrations were determined by using samples collected with a Harvard impactor. However, sampling duration was 240 minutes, due to concern about collecting sufficient amount of $PM_{2.5}$ for weighing. As a result, the change in

concentration from background to during frying could not be differentiated. In Table 4.7 summary statistics of $PM_{2.5}$ concentrations are presented. The mean $PM_{2.5}$ concentration of three days sampling was $108~\mu g/m^3$, and the median $PM_{2.5}$ concentration was $90.0~\mu g/m^3$.

Table 4.7. Summary Statistics of $PM_{2.5}$ Concentrations ($\mu g/m^3$) in the Restaurant Kitchen(n=3)

Mean	108
Median	90.0
SD	44.2
Min	75.7
Max	158

4.2. Campaign-1, Monitoring

In this section, results of the three-day measurement in Campaign-1 are presented for PM_{10} , TVOC, CO_2 , CO, temperature and RH.

4.2.1. PM₁₀ Concentrations

 PM_{10} concentrations are presented as five-minute average values along with corresponding standard deviations in Figure 4.3. Before frying period is the first 60 minutes. Five-minute mean PM_{10} concentrations were ranged from 332 $\mu g/m^3$ to 1104 $\mu g/m^3$ in this period. Median concentrations were ranged from 359 $\mu g/m^3$ to 486 $\mu g/m^3$. During frying period (shown as gray shaded area) was from the 90^{th} minute to the 150^{th} minute. Mean PM_{10} concentrations were ranged from 694 $\mu g/m^3$ to $1583 \mu g/m^3$. Median concentrations were ranged from 356 $\mu g/m^3$ to $1606 \mu g/m^3$ Minimum and maximum concentrations were measured as 277 $\mu g/m^3$ and 3691 $\mu g/m^3$, respectively. In after frying period (180^{th} minute to 240^{th} minute), minimum PM_{10} concentration was 32.0 $\mu g/m^3$ and maximum PM_{10} concentration was recorded as $1649 \mu g/m^3$ with the mean and median concentration ranges were from 259 $\mu g/m^3$ to 831 $\mu g/m^3$ and from 226 $\mu g/m^3$ to 779 $\mu g/m^3$, respectively. The increase in the mean concentration from the before to the during frying periods was 2 folds.

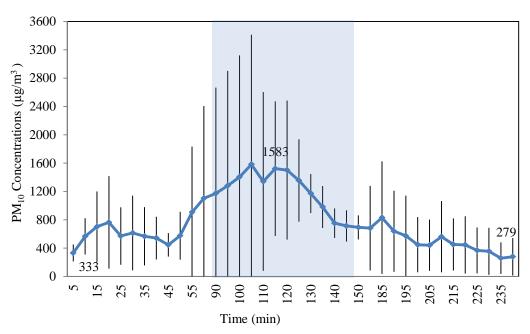


Figure 4.3 Campaign-1, Monitoring Mean PM₁₀ Concentrations

4.2.2. TVOC Concentrations

Summary statistics of TVOC concentrations are presented as five-minute average values along with corresponding standard deviations in Figure 4.4. In first 60 minutes period which is before frying period, the mean concentrations ranged from 11.4 ppb to 39.2 ppb. During frying period was from the 90th minute to the 150th minute for which minimum and maximum concentrations were 4.37 ppb and 91.0 ppb, respectively. Mean TVOC concentration was ranged from 11.0 ppb to 60.7 ppb. In after frying period which was from the 180th minute to the 240th minute, mean TVOC concentrations were ranged from 9.51 ppb to 21.5 ppb. The increase in mean concentrations were ranged from the before to the during frying period was 1.5 folds.

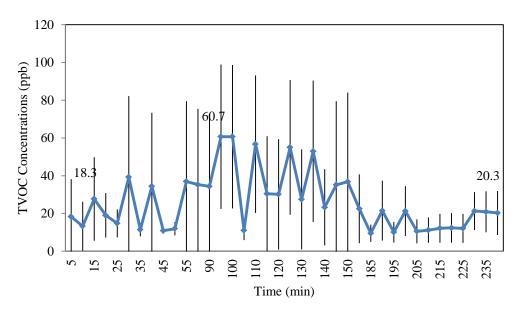


Figure 4.4. Campaign-1, Monitoring Mean TVOC Concentrations

4.2.3. CO₂ Concentrations

Mean CO₂ concentrations were measured in the range from 537 ppm to 648 ppm in before frying period, from 532 ppm to 559 ppm in during frying period and from 433 ppm to 470 ppm in after frying period. Also, maximum CO₂ concentrations were determined as 792 ppm, 620 ppm and 577 ppm for before frying, during frying and after frying periods, respectively. CO₂ concentrations were seemed as not to be related to the frying activity as the fryer was electric and only one or two persons were present in the kitchen.

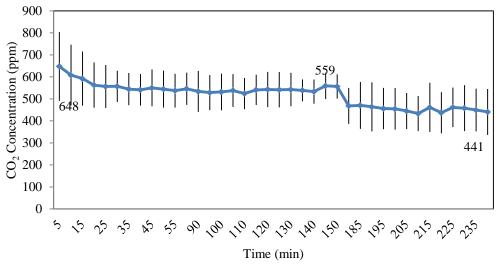


Figure 4.5. Campaign-1, Monitoring Mean CO₂ Concentrations

4.2.4. CO Concentrations

Mean CO concentrations were ranged between 2.05 ppm and 2.67 ppm in before frying period, 2.12 ppm and 2.67 ppm in during frying period and lastly 2.73 ppm and 3.63 ppm in after frying period. Also, maximum CO concentrations were determined as 4.00 ppm, 4.60 ppm and 7.10 ppm for before frying, during frying and after frying periods respectively. In Figure 4.6 mean CO concentrations are shown. There seems to be a faint relation between CO concentrations and the frying as the concentrations and the frying as the concentrations were increased somewhat towards the end of the measurement period, indicating a build up. However, this delayed increase in the concentrations may also not be related to frying.

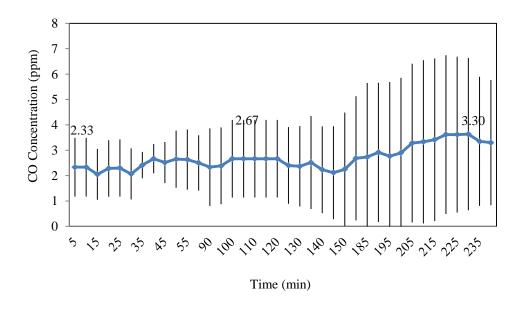


Figure 4.6. Campaign-1, Monitoring Mean CO Concentrations

4.2.5. Temperature

Due to a device realted problem data for two days were lost. Therefore, temperatures for only one day are presented in Figure 4.7. The results showed that temperature was constant at 27 °C except before frying period, temperature was ranged from 23 °C to 27 °C. The increase was probably related to th increase in the ooutdoor temparature during the day.

4.2.6. Relative Humidity

Due to the same problem regarding temperature only one day RH levels are presented in Figure 4.8. According to the results in before frying period, RH levels were ranged from 46% to 51%. In during frying and after frying periods RH levels were ranged from 41% to 44% and 39% to 41% respectively.

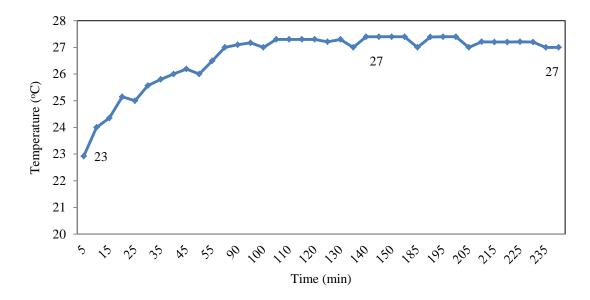


Figure 4.7. Campaign-1, Monitoring Temperature Levels

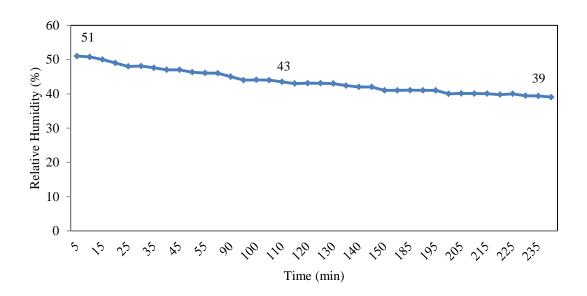


Figure 4.8. Campaign-1, Monitoring Relative Humidity Levels

4.3. Campaign-2, Sampling

In the second campaign of the study which was started on 22nd of January and ended on 7th of February, sampling and measurements were performed under controlled conditions for three days in the same restaurant kitchen. Only French cut potatoes were fried with the vegetable margarine in the deep fryer. The amount of fried potatoes in one hour period was varied as 1250 g, 2500 g, and 3750 g in the first, second, and third day of the campaign, respectively.

4.3.1. VOC Concentrations

47 target VOC compounds were detected in all sampling periods in the restaurant kitchen except for 2-butanone which was not detected in some samples. Summary statistics of the measured VOC concentrations with varying amount of potatoes fried are shown in Table 4.8, Table 4.9 and Table 4.10 for before frying sampling, during frying sampling and after frying sampling, respectively.

Concentrations of 35 VOCs were increased in during frying sampling period compared to the background levels, which were ethyl acetate, 4-methyl 2-pentanone, toluene, n-octane, n-butyl acetate, tetrachloroethene, ethyl benzene, p/m xylene, styrene, o-xylene, 1,3,5 trimethyl benzene, β-pinene, 2 ethyl toluene, limonene, 1,2,3 trimethylbenzene, n-undecane, nonanal, 1,2,4,5-tetramethylbenzene, n-dodecane, decanal, n-tridecane, n-tetradecane, n-pentadecane, n-hexadecane, 1,1,1-trichloroethane, 1,2-dichloroethane, n-butanol, carbontetrachloride, iso-octane, n-heptane, trichloroethylene, 1,2-dichloropropane, bromodichloromethane, 2-propanol, 1-propanol. In the first campaign of the study, 22 VOCs increased in during frying period 17 of them are the same VOCs in the second campaign of the study, including the four compounds that increased the most. TVOC mean concentrations which were determined by the sum of detected VOCs were 140 μg/m³, 155 μg/m³, and 106 μg/m³ for before frying, during frying, and after frying sampling periods, respectively.

Table 4.8. Summary Statistics of Before Frying Period VOC Concentrations ($\mu g/m^3$) in the Restaurant Kitchen

True of Commence de		2 nd Day	ard D	1.7	N / 1º	CD
Target Compounds	1 st Day	/	3 rd Day	Mean	Median	SD
1,2,4-trimethylbenzene	0.84	0.37	1.09	0.77	0.84	0.37
ethyl acetate	1.09	22.56	0.53	8.06	1.09	12.56
4 methyl 2 pentanone	N.D.	0.06	0.07	0.07	0.07	0.00
toluene	2.45	5.73	1.92	3.37	2.45	2.06
n-octane	9.46	1.71	5.80	5.66	5.80	3.88
dibromochloromethane	0.02	0.01	0.02	0.02	0.02	0.01
n-butyl acetate	0.17	0.77	0.18	0.37	0.18	0.34
tetrachloroethene	0.04	0.37	0.03	0.15	0.04	0.19
ethylbenzene	0.43	0.55	0.44	0.47	0.44	0.07
p-m-xylene	0.57	0.86	0.58	0.67	0.58	0.16
n-nonane	2.31	0.41	2.67	1.80	2.31	1.21
styrene	0.08	0.53	0.07	0.23	0.08	0.26
o-xylene	0.50	0.63	0.53	0.55	0.53	0.07
α-pinene	0.53	2.11	1.47	1.37	1.47	0.79
3-ethyltoluene	0.53	0.25	0.67	0.48	0.53	0.21
4-ethyltoluene	0.22	0.10	0.27	0.20	0.22	0.09
1,3,5-trimethylbenzene	0.20	0.09	0.29	0.19	0.20	0.10
n-decane	2.59	0.61	2.62	1.94	2.59	1.15
β-pinene	0.10	0.33	1.23	0.55	0.33	0.60
2-ethyltoluene	0.17	0.05	0.24	0.15	0.17	0.10
1,4-dichlorobenzene	37.3	1.00	27.3	21.9	27.3	18.7
limonene	0.50	0.94	4.23	1.89	0.94	2.04
1,2,3-trimethylbenzene	0.23	0.06	0.34	0.21	0.23	0.14
n-undecane	1.04	0.03	1.19	0.75	1.04	0.63
nonanal	19.5	6.73	12.0	12.8	12.0	6.43
1,2,4,5-tetramethylbenzene	0.02	0.03	0.12	0.06	0.03	0.05
n-dodecane	1.14	0.18	1.05	0.79	1.05	0.53
decanal	4.54	1.25	2.54	2.77	2.54	1.66
n-tridecane	0.86	0.20	0.76	0.60	0.76	0.36
n-tetradecane	0.85	0.39	0.76	0.67	0.76	0.24
n-pentadecane	0.71	0.28	0.59	0.53	0.59	0.22
n-hexadecane	0.53	0.22	0.41	0.39	0.41	0.16
hexane	0.71	0.22	0.41	0.53	0.41	0.10
2-butanone	BDL	BDL	0.09	0.09	0.09	BDL
chloroform	0.49	0.29	0.39	0.09	0.39	0.10
1,1,1-trichloroethane	0.49	0.29	0.02	0.03	0.39	0.10
1,2-dichloroethane						
,	0.05	0.85	0.07	0.32	0.07	0.45
n-butanol	0.78	0.84	0.48	0.70	0.78	0.19
carbontetrachloride	0.57	0.51	0.50	0.53	0.51	0.04
benzene	1.55	4.08	3.56	3.06	3.56	1.34
iso-octane	0.76	0.71	0.61	0.69	0.71	0.08

Table 4.8 (cont.)

Target Compounds	1 st Day	2 nd Day	3 rd Day	Mean	Median	SD
n-heptane	44.83	4.89	23.9	24.6	23.9	20.0
tricloroethylene	0.01	0.99	0.01	0.34	0.01	0.57
1,2-dichloropropane	0.02	0.02	0.02	0.02	0.02	0.00
bromodichloromethane	0.03	0.03	0.03	0.03	0.03	0.00
2-propanol	0.13	0.19	0.04	0.12	0.13	0.07
methylene chloride	0.05	0.39	BDL	0.22	0.22	0.24
1-propanol	0.16	0.04	0.04	0.08	0.04	0.07
ΣΤVΟC	140	63.9	102	102	102	37.9

Table 4.9. Summary Statistics of During Frying Period VOC Concentrations $(\mu g/m^3)$ in the Restaurant Kitchen

Target Compounds	1 st Day	2 nd Day	3 rd Day	Mean	Median	SD
1,2,4-trimethylbenzene	1.25	0.27	0.94	0.82	0.94	0.50
ethyl acetate	1.61	7.13	1.89	3.54	1.89	3.11
4 methyl 2 pentanone	0.18	0.09	0.14	0.14	0.14	0.05
toluene	3.59	3.24	1.98	2.93	3.24	0.85
n-octane	13.5	6.94	16.7	12.4	13.5	4.98
dibromochloromethane	0.02	0.01	0.02	0.02	0.02	0.00
n-butyl acetate	0.24	0.38	0.19	0.27	0.24	0.10
tetrachloroethene	0.05	0.18	0.05	0.09	0.05	0.08
ethylbenzene	0.71	0.32	0.45	0.49	0.45	0.20
p-m-xylene	0.91	0.43	0.56	0.63	0.56	0.25
n-nonane	3.54	0.71	2.40	2.21	2.40	1.42
styrene	0.12	0.13	0.07	0.11	0.12	0.03
o-xylene	0.80	0.32	0.48	0.54	0.48	0.24
α-pinene	0.64	1.69	1.70	1.34	1.69	0.61
3-ethyltoluene	0.79	0.19	0.57	0.52	0.57	0.30
4-ethyltoluene	0.33	0.07	0.22	0.21	0.22	0.13
1,3,5-trimethylbenzene	0.32	0.07	0.22	0.21	0.22	0.13
n-decane	3.48	1.13	3.04	2.55	3.04	1.25
β-pinene	0.24	0.17	0.65	0.35	0.24	0.26
2-ethyltoluene	0.29	0.10	0.20	0.20	0.20	0.10
1,4-dichlorobenzene	23.68	4.95	29.63	19.4	23.7	12.9
limonene	0.83	1.06	2.82	1.57	1.06	1.09
1,2,3-trimethylbenzene	0.38	0.09	0.28	0.25	0.28	0.15
n-undecane	1.77	0.10	1.54	1.13	1.54	0.90
nonanal	20.0	13.1	25.2	19.4	20.0	6.05
1,2,4,5-tetramethylbenzene	0.07	0.02	0.04	0.04	0.04	0.02
n-dodecane	1.63	0.27	1.33	1.08	1.33	0.72
decanal	3.61	2.66	3.57	3.28	3.57	0.54
n-tridecane	1.23	0.26	0.95	0.81	0.95	0.50
n-tetradecane	1.13	0.53	1.17	0.94	1.13	0.36

Table 4.9. (cont.)

Target Compounds	1 st Day	2 nd Day	3 rd Day	Mean	Median	SD
n-pentadecane	1.04	0.55	1.37	0.99	1.04	0.41
n-hexadecane	0.65	0.34	0.57	0.52	0.57	0.16
hexane	0.69	0.60	0.53	0.61	0.60	0.08
2-butanone	BDL	0.04	0.03	0.04	0.04	< 0.01
chloroform	0.21	0.21	0.66	0.36	0.21	0.26
1,1,1-trichloroethane	0.03	0.03	0.03	0.03	0.03	0.00
1,2-dichloroethane	0.08	0.72	0.06	0.29	0.08	0.38
n-butanol	1.43	0.72	1.64	1.27	1.43	0.48
carbontetrachloride	0.50	0.48	0.61	0.53	0.50	0.07
benzene	1.58	2.80	1.70	2.03	1.70	0.67
iso-octane	1.05	0.51	0.81	0.79	0.81	0.27
n-heptane	60.2	30.8	80.7	57.3	60.2	25.1
tricloroethylene	0.01	0.35	0.02	0.13	0.02	0.19
1,2-dichloropropane	0.02	0.02	0.03	0.02	0.02	0.00
bromodichloromethane	0.03	0.03	0.04	0.03	0.03	0.01
2-propanol	0.15	1.12	0.13	0.47	0.15	0.57
methylene chloride	0.03	0.09	0.14	0.09	0.09	0.05
1-propanol	0.15	0.10	0.20	0.15	0.15	0.05
ΣΤΥΟС	155	86.2	188	143	155	52.1

BDL: Below Detection Limit

Table 4.10. Summary Statistics of After Frying Period VOC Concentrations ($\mu g/m^3$) in the Restaurant Kitchen

Target Compounds	1 st Day	2 nd Day	3 rd Day	Mean	Median	SD
1,2,4-trimethylbenzene	1.08	0.32	0.87	0.76	0.87	0.39
ethyl acetate	1.28	2.26	2.58	2.04	2.26	0.68
4 methyl 2 pentanone	0.06	0.07	0.09	0.07	0.07	0.02
toluene	3.16	10.46	6.71	6.78	6.71	3.65
n-octane	6.09	2.47	6.29	4.95	6.09	2.15
dibromochloromethane	0.02	0.01	0.03	0.02	0.02	0.01
n-butyl acetate	0.21	2.07	0.12	0.80	0.21	1.10
tetrachloroethene	0.05	0.05	0.04	0.05	0.05	0.01
ethylbenzene	0.60	0.24	0.82	0.55	0.60	0.29
p-m-xylene	0.78	0.27	1.67	0.91	0.78	0.71
n-nonane	2.75	0.68	1.66	1.70	1.66	1.03
styrene	0.10	0.06	0.14	0.10	0.10	0.04
o-xylene	0.69	0.23	0.76	0.56	0.69	0.29
α-pinene	0.87	1.36	1.10	1.11	1.10	0.25
3-ethyltoluene	0.70	0.22	0.60	0.51	0.60	0.25
4-ethyltoluene	0.29	0.08	0.31	0.23	0.29	0.13
1,3,5-trimethylbenzene	0.28	0.09	0.20	0.19	0.20	0.09

Table 4.10. (cont.)

Target Compounds	1 st Day	2 nd Day	3 rd Day	Mean	Median	SD
n-decane	2.80	0.72	2.13	1.89	2.13	1.06
β-pinene	1.57	0.10	0.25	0.64	0.25	0.81
2-ethyltoluene	0.26	0.10	0.15	0.17	0.15	0.08
1,4-dichlorobenzene	20.2	35.9	27.4	27.8	27.4	7.84
limonene	3.10	0.29	1.24	1.54	1.24	1.43
1,2,3-trimethylbenzene	0.32	0.09	0.18	0.20	0.18	0.11
n-undecane	1.30	0.10	0.70	0.70	0.70	0.60
nonanal	16.3	10.3	20.2	15.6	16.3	4.98
1,2,4,5-tetramethylbenzene	0.05	0.03	0.08	0.05	0.05	0.02
n-dodecane	1.42	0.27	0.91	0.87	0.91	0.58
decanal	4.07	1.93	4.49	3.50	4.07	1.37
n-tridecane	1.02	0.27	0.73	0.68	0.73	0.38
n-tetradecane	0.96	0.44	0.79	0.73	0.79	0.27
n-pentadecane	0.86	0.38	0.78	0.67	0.78	0.26
n-hexadecane	0.60	0.34	0.51	0.48	0.51	0.13
hexane	0.54	0.25	2.92	1.24	0.54	1.46
2-butanone	BDL	BDL	< 0.01	< 0.01	< 0.01	BDL
chloroform	0.19	0.44	0.69	0.44	0.44	0.25
1,1,1-trichloroethane	0.03	0.04	0.04	0.04	0.04	0.00
1,2-dichloroethane	0.04	0.36	0.06	0.15	0.06	0.18
n-butanol	0.70	0.72	0.60	0.67	0.70	0.07
carbontetrachloride	0.53	0.58	0.79	0.63	0.58	0.13
benzene	1.41	1.69	7.21	3.44	1.69	3.27
iso-octane	0.94	0.45	0.86	0.75	0.86	0.26
n-heptane	27.4	10.3	26.6	21.4	26.6	9.66
tricloroethylene	0.00	0.00	0.01	0.01	0.00	0.01
1,2-dichloropropane	0.02	0.02	0.02	0.02	0.02	0.00
bromodichloromethane	0.02	0.04	0.04	0.03	0.04	0.01
2-propanol	0.16	2.69	0.16	1.00	0.16	1.46
methylene chloride	0.05	0.16	0.12	0.11	0.12	0.06
1-propanol	0.08	< 0.01	0.14	0.07	0.08	0.07
ΣΤVΟC	106	89.9	125	107	106	17.5

BDL: Below Detection Limit

Mean concentrations of eight VOCs which increased the most in during frying sampling period are presented in Figure 4.9. The VOCs were n-heptane, nonanal, n-octane, ethyl acetate, limonene, n-butanol, n-undecane and n-dodecane. Four of the eight VOCs which are n-heptane, nonanal, n-octane, ethyl acetate are the same with the most increased the ones in the first campaign of the study. The mean concentration of n-heptane, nonanal, n-octane, ethyl acetate in the first campaign of the study was higher than the mean concentrations in the second campaign of the study. The difference is

probably related to frying of hamburger patties, chicken patties, nuggets and schnitzels that were among the fired items in Campaign-1.

In addition, the concentration increase in the eight VOCs with the increasing amount of fried potatoes is presented in Figure 4.10 which shows that the relation between the two is apparent for n-heptane, nonanal, and n-octane.

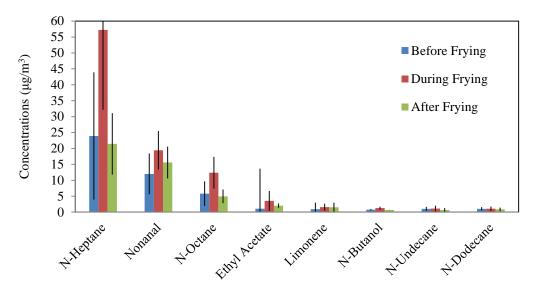


Figure 4.9. VOCs Concentrations with the Highest Increase in During Frying Period in the Restaurant Kitchen

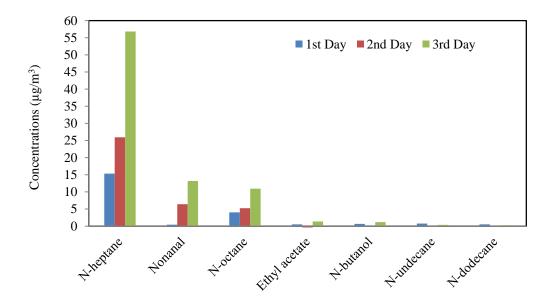


Figure 4.10. Increased Amount of VOC Concentrations in the Restaurant Kitchen

4.3.2. Aldehyde Concentrations

In this campaign, 12 aldehydes were detected in all periods of the study. Detected aldehydes were formaldehyde, acetaldehyde, acetone, propionaldehyde, butryaldehyde, benzaldehyde, valeraldehyde, o-tolualdehyde, m/p-tolualdehyde, hexaldehyde, 2.5-dimethylbenzaldehyde. Crotonaldehyde, isovaleraldehyde, were not detected in all periods of sampling. Summary statistics of the measured aldehyde concentrations with varying amount of potatoes fried are shown in Table 4.11, Table 4.12 and Table 4.13 for before frying sampling, during frying sampling, and after frying sampling, respectively. Total aldehydes concentrations were 5.24, 4.83, 3.85 μ g/m³ for the first day, 2.65, 4.35, 5.18 μ g/m³ for the second day, and 4.26 , 4.09, 2.68 μ g/m³ for the third day of the campaign in before frying, during frying, and after frying periods, respectively.

Table 4.11. Summary Statistics of Before Frying Period Aldehyde Concentrations $(\mu g/m^3)$ in the Restaurant Kitchen

(µg/m / m the Re				1		
Target Compound	1 st Day	2 nd Day	3 rd Day	Mean	Median	SD
formaldehyde	2.33	1.43	1.79	1.85	1.79	0.45
acetaldehyde	0.40	0.54	0.86	0.60	0.54	0.24
propionaldehyde	0.33	0.15	0.54	0.34	0.33	0.20
crotonaldehyde	0.00	0.00	0.00	0.00	0.00	0.00
butryaldehyde	0.26	0.16	0.16	0.20	0.16	0.06
benzaldehyde	0.17	0.06	0.09	0.10	0.09	0.06
isovaleraldehyde	0.13	0.00	0.00	0.04	0.00	0.08
valeraldehyde	0.41	0.11	0.20	0.24	0.20	0.16
o-tolualdehyde	0.00	0.00	0.00	0.00	0.00	0.00
m/p-tolualdehyde	0.08	0.03	0.06	0.05	0.06	0.03
hexaldehyde	1.02	0.01	0.56	0.53	0.56	0.51
2,5-dimethylbenzaldehyde	0.10	0.18	0.00	0.10	0.10	0.09
total aldehydes	5.24	2.65	4.26	4.05	4.26	1.31

Table 4.12. Summary Statistics of During Frying Period Aldehyde Concentrations $(\mu g/m^3)$ in the Restaurant Kitchen

(µg/m/) m the rest		nd nd	rd ed	1	1	
Target Compound	1 st Day	2 nd Day	3 rd Day	Mean	Median	SD
formaldehyde	1.63	1.98	1.08	1.56	1.63	0.45
acetaldehyde	0.76	0.91	0.88	0.85	0.88	0.08
propionaldehyde	0.27	0.22	0.24	0.24	0.24	0.02
crotonaldehyde	0.00	0.00	0.00	0.00	0.00	0.00
butryaldehyde	0.26	0.27	0.24	0.26	0.26	0.02
benzaldehyde	0.20	0.13	0.13	0.15	0.13	0.04
isovaleraldehyde	0.00	0.00	0.00	0.00	0.00	0.00
valeraldehyde	0.42	0.21	0.31	0.32	0.31	0.10
o-tolualdehyde	0.00	0.00	0.00	0.00	0.00	0.00
m/p-tolualdehyde	0.05	0.05	0.07	0.06	0.05	0.01
hexaldehyde	1.04	0.52	1.00	0.86	1.00	0.29
2,5-dimethylbenzaldehyde	0.19	0.05	0.14	0.13	0.14	0.07
total aldehydes	4.83	4.35	4.09	4.42	4.35	0.37

Table 4.13. Summary Statistics of After Frying Period Aldehyde Concentrations $(\mu g/m^3)$ in the Restaurant Kitchen

Target Compound	1 st Day	2 nd Day	3 rd Day	Mean	Median	SD
formaldehyde	1.63	3.21	1.06	1.97	1.63	1.12
acetaldehyde	0.47	0.64	0.62	0.58	0.62	0.09
propionaldehyde	0.34	0.17	0.22	0.24	0.22	0.09
crotonaldehyde	0.00	0.00	0.00	0.00	0.00	0.00
butryaldehyde	0.15	0.26	0.03	0.15	0.15	0.12
benzaldehyde	0.16	0.13	0.12	0.14	0.13	0.02
ısovaleraldehyde	0.22	0.00	0.00	0.07	0.00	0.13
valeraldehyde	0.13	0.14	0.16	0.14	0.14	0.01
o-tolualdehyde	0.00	0.00	0.00	0.00	0.00	0.00
m/p-tolualdehyde	0.05	0.07	0.04	0.05	0.05	0.02
hexaldehyde	0.61	0.52	0.39	0.50	0.52	0.11
2,5-dimethylbenzaldehyde	0.07	0.02	0.04	0.05	0.04	0.03
total aldehydes	3.85	5.18	2.68	3.90	3.85	1.25

In Campaign-2, aldehydes with the highest increase in concentrations in during frying period were Hexaldehyde, Acetaldehyde, Valeraldehyde, Butryaldehyde, Benzaldehyde, 2,5-Dimethylbenzaldehyde (Figure 4.11)

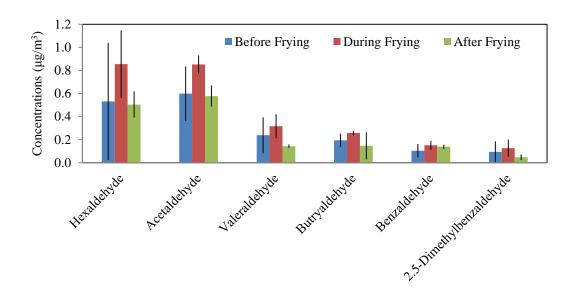


Figure 4.11. Aldehyde Concentrations with Highest Increase in During Frying Period in the Restaurant Kitchen

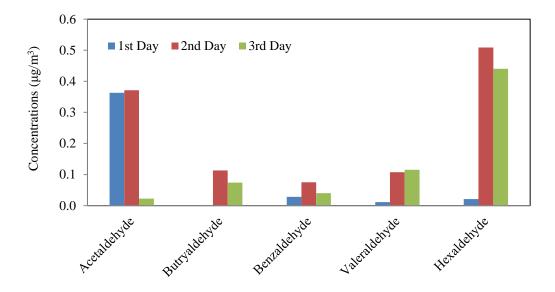


Figure 4.12. Increased Amount of Aldehyde Concentrations in the Restaurant Kitchen

In addition, the increase concentration in the six aldehydes with the increasing amount of fried potatoes presented in Figure 4.12, which shows that the relation between two is not apparent for target aldehydes.

4.3.3. PM_{2.5} Concentrations

Due to detection limit concerns, before, during, after frying differentiation could not be made, $PM_{2.5}$ concentration was determined as 4-hour average. Therefore, there is one concentration for each of the sampling days in the campaign. Table 4.14 lists the measured concentrations, along with the mean and median values. The increase in the 4-hr average $PM_{2.5}$ concentration indicates that emission of particles is related to the amount of potatoes fried (Figure 4.13).

Table 4.14. Summary Statistics of $PM_{2.5}$ Concentrations ($\mu g/m^3$) in the Restaurant Kitchen (n=3)

	$PM_{2.5} (\mu g/m^3)$
1st Day	81.3
2nd Day	110
3rd Day	249
Mean	147
Median	110
SD	89.8
Min	81.3
Max	249

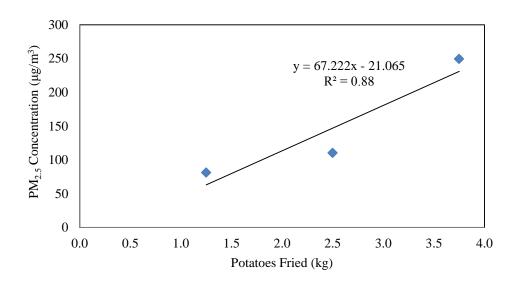


Figure 4.13 Simple Liner Regression of PM_{2.5} and Amount of Potatoes Fried

4.4. Campaign-2, Monitoring

4.4.1. PM₁₀ Concentrations

Before frying sampling results are presented in first 60 minutes. Different from Campaign-1, since this was a controlled experiment, there was no frying activity in this period. As a result, it can be seen that the concentrations were fairly constant during this period (Figure 4.14). Mean concentrations were ranged from 334 μ g/m³ to 769 μ g/m³ in the before frying period. The during frying period was started in the 90th minute and ended in the 150th minute. In the first day of the study, PM₁₀ concentrations were ranged from 724 μ g/m³ to 3292 μ g/m³. In the second day, PM₁₀ concentrations were ranged from 324 μ g/m³ to 5554 μ g/m³ and in the third day of the study, PM₁₀ concentrations were ranged from 1814 μ g/m³ 9070 μ g/m³ for 1250 g, 2500 g and 3750 g french fries respectively. Also, mean of three day PM₁₀ concentrations were ranged from 964 μ g/m³ to 5779 μ g/m³. After frying period was from 180th minute to the 240th minute. Mean PM₁₀ concentration levels were measured from 231 μ g/m³ to 1358 μ g/m³.

Figure 4.14 shows the first day, second day, third day, and overall mean PM_{10} concentration variation in the before, during, and after frying periods. In the before frying period PM_{10} concentrations were low; with the beginning of frying activity PM_{10} concentrations started to increase, reaching its maximum level at 115^{th} minute as $3292 \, \mu g/m^3$, at 130^{th} minute as $5554 \, \mu g/m^3$, and at 140^{th} minute as $9070 \, \mu g/m^3 \, m^3$ in the first, second, and third day of the campaign, respectively. The time lag in the peak concentration should be related to the amount of time that takes to fry the increasing amount of potatoes. The highest mean concentration was measured as $5779 \, \mu g/m^3$ in during frying period. PM_{10} concentrations started to decrease in the after frying period, and it reached its lowest level at the end of the monitoring period in all measurement days. In addition, to determine the effects of amount of potatoes fried PM_{10} versus amount of potatoes fried graph was plotted. The increase in the average PM_{10} concentration indicates that emission of particles is related to the amount of potatoes fried (Figure 4.15).

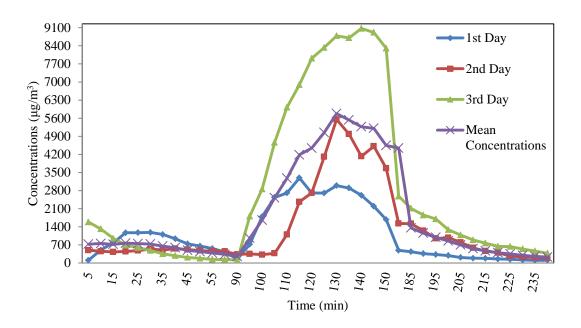


Figure 4.14. Campaign-2 Monitoring PM_{10} Concentration

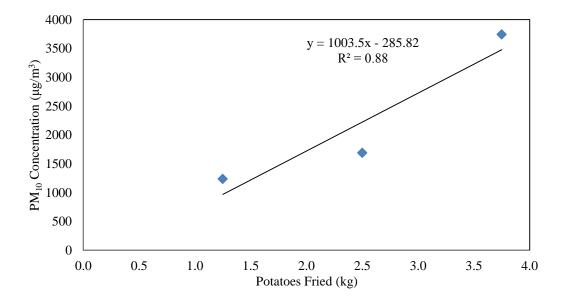


Figure 4.15. Simple Linear Regression of PM₁₀ and Amount of Potatoes Fried

4.4.2. TVOC Concentrations

Five minute average TVOC concentrations are shown against time in Figure 4.16 for the three monitoring days and as the three-day averages. The mean concentrations ranged from 9.60 ppb to 31.8 ppb in the before frying period (the first 60

mins). The during frying period was from the 90th minute to the 150th minute. The first day, TVOC concentrations were ranged from 8.13 ppb to 11.5 ppb in during frying period. The second day concentrations were ranged from 6.36 ppb to 10.4 ppb, and the third day TVOC levels were ranged from 5.55 ppb to 8.74 ppb in the during frying period. The mean TVOC concentration was ranged from 7.69 ppb to 9.60 ppb in during frying period. In the after frying period, which was from the 180th minute to the 240th minute, mean TVOC concentrations were ranged from 5.32 ppb to 6.51 ppb. TVOC concentrations were not affected by the increasing amount of potatoes in contrast to the PM concentrations. Actually, TVOC levels were higher in the before period compared to the during and after periods. The decrease may have occurred due to sorption on the particles which were increasing due to the frying.

When we compare the results with Campaign-1, TVOC concentrations in the second campaign were lower than the first campaign TVOC concentration levels. A possible explanation may be, when frozen meat products which were schnitzels, nuggets, hamburger patties, and chicken patties are fried with the vegetable margarine, relatively higher VOC emissions occur.

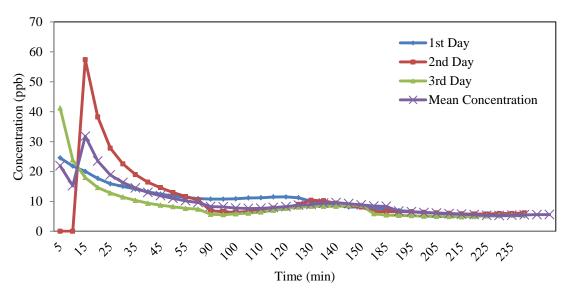


Figure 4.16. Campaign-2 Monitoring TVOC Concentration

4.4.3. CO₂ Concentrations

When we look at the CO₂ concentration levels in the second campaign of the study in before frying period, mean CO₂ concentrations were ranged from 457 ppm to

713 ppm. In during frying period, first day CO₂ concentrations were ranged from 362 ppm to 428 ppm. CO₂ levels ranged from 423 ppm to 464 ppm for second day and third day CO₂ results were between 463 ppm and 547 ppm levels. In after frying period, mean CO₂ concentrations were ranged from 423 ppm to 487 ppm. Five minute average CO₂ concentrations are shown against time in Figure 4.17 for the three monitoring days and as the three-day averages.

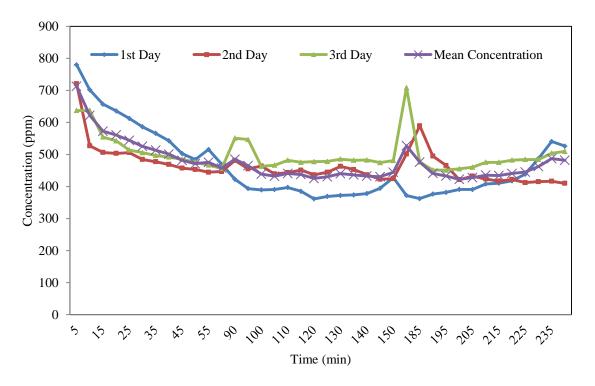


Figure 4.17. Campaign-2 Monitoring CO₂ Concentration

4.4.4. CO Concentrations

Five minute average CO concentrations are shown against time in Figure 4.18 for the three monitoring days and as the three-day averages. In the first day of campaign before frying CO levels were ranged from 0 ppm to 0.5 ppm. In during frying and after frying period CO was not detected. In second day, before frying CO concentrations were detected at 1 ppm approximately. The levels of CO in during frying period were ranged from 0.25 ppm to 1 ppm. In after frying period maximum detected CO level was 0.40 ppm. In the third day of the campaign, before frying CO levels equal to 1 ppm approximately. In during frying and after frying periods CO was not detected. The concentration profile is very different than that was observed in Campaign-1 which may

indicate that the delayed CO increase in the 1st campaign was probably not related to frying.

4.4.5. Temperature

Five minute average temperature levels are shown in Figure 4.19. In the restaurant kitchen all windows were closed during measurements and because of during frying times were at lunch time the temperature results increased in during frying periods. In before frying period mean temperature levels were ranged from 21 °C to 26 °C. In during frying period, mean temperature levels were ranged from 26 °C to 27 °C. Lastly, in after frying period, mean temperature levels were detected in range from 25 °C to 27 °C. The temperature profile is smilar to that of observed in Campaign-1.

4.4.6. Relative Humidity

Five minute average RH levels are shown in Figure 4.20. In before frying period, mean levels of relative humidity were ranged from 45 % to 52 %. In during frying period mean realtive humidity levels were detected between 43 % and 44 %. In after frying period, relative humidity levels were ranged from 42 % to 44 %. The RH profile is smilar to that of observed in Campaign-1.

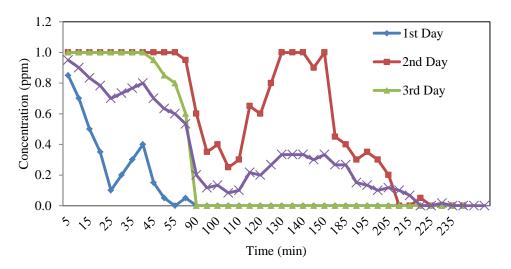


Figure 4.18. Campaign-2 Monitoring CO Concentration

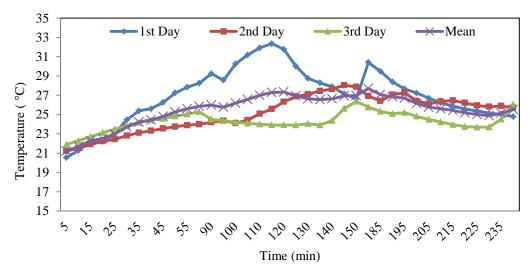


Figure 4.19. Campaign-2 Monitoring Temperature Levels

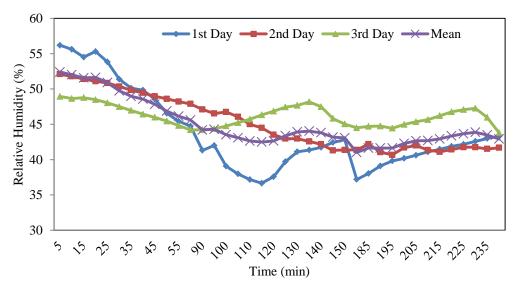


Figure 4.20. Campaign-2 Monitoring Relative Humidity Levels

CHAPTER 5

DISCUSSION

The main goal of the experiments conducted in this study was to determine indoor air pollutant levels in a small scale restaurant kitchen where french fries and other foods were fried in deep fryers with a special vegetable margarine for frying to estimate cooks exposure during frying activities in the kitchen. To do this, we sampled and measured indoor air pollutants in a small scale restaurant kitchen in two campaigns: (1) real life conditions and (2) under controlled conditions.

In this section, we compared the results of sampled and measured indoor air pollutants that were detected kitchen with the literature and standard levels. Indoor concentrations in restaurant or house kitchens usually exceed outdoor concentration levels (Abt et al., 2000; Lee et al., 2001; Fortmann et al., 2001; Lee et al., 2002). Vainiotalo and Matveinen, 1993; Thiebaud et al., 1995; Gertz, 2000; Fortman et al., 2001; Svendsen et al., 2002; Fullana et al., 2004a,b; See et al., 2006; Zhao et al., 2007; Sjaastad et al., 2008; See and Balasubramanian, 2008; Katragadda et al., 2010 have studied levels of indoor air pollutants in the breathing zone of the cook, investigating with different methods, frying foods, oil types, frying methods etc. In addition to all of the above parameters presence of ventilation hoods and exhausts system makes it difficult to make direct comparisons of the exposure concentrations measured in this study.

5.1. VOC Concentrations

The samples were analyzed for a total of 48 VOCs in this study. Except one of them which was 2-butanone, 47 target VOC compounds were detected in all sampling periods in the restaurant kitchen. In the first campaign of the study, concentrations of 22 VOCs were increased, in the second campaign concentrations of 35 VOCs were increased during frying compared to the background. Seventeen of the increased VOCs in the first campaign were the same VOCs that increased in the second campaign of the study. VOCs with the highest average indoor concentrations were varied in during

frying sampling period probably depending on the foods that were fried and their amounts. In the first campaign of the study, VOCs with the highest mean indoor concentrations were n-heptane, ethyl acetate, nonanal, n-octane, toluene, decanal, ndecane and hexane with 83.0, 24.9, 23.4, 16.6, 4.37, 3.12, 1.91, 0.90 μ g/m³, respectively. In the second campaign of the study VOCs with the highest mean indoor concentrations were n-heptane, nonanal, n-octane, ethyl acetate, limonene, n-butanol, nundecane and n-dodecane with 57.3, 19.4, 12.4, 3.5, 1.57, 1.27, 1.13 and 1.08 μg/m³, respectively. In addition, TVOC concentrations were calculated by the sum of determined VOCs concentrations and also TVOC concentration levels were measured with continuous air monitoring device. TVOC concentrations were determined as for the first and the second campaign of the study as 207 µg/m³ and 155 µg/m³ respectively by sum of determined VOCs concentrations. Mean concentration range of TVOC levels were determined with the continuous air monitoring device in the first and the second campaign of the study during frying as from 11.0 ppb to 56.7 ppb and from 7.69 ppb to 9.60 ppb, respectively. There is a discrepancy between the sampling TVOC and monitoring TVOC concentrations if we assume a 1/2.3 for (ppb)/(µg/m³) conversion (Hong Kong, 2003). Since sampling method is more dependable monitoring TVOC levels should be taken with care. We used the monitoring TVOC to investigate fluctuations in before, during and after frying periods.

In literature, there are various studies that were determined indoor concentrations of poliaromatic hydrocarbons (PAHs) and aldehydes in restaurant kitchens during frying of various food types (Svendsen et al., 2002; Fortmann et al., 2002; Kelly, 2001; Wu et al., 1998; Zhu and Wang, 2003; Li et al., 1994; Boskou et al., 2006; Zhao et al., 2007a,b; Vainiotalo and Matveinen, 1993) however, there are few studies that have studied VOC (alkanes, aliphatic hydrocarbons, alkanones, terpenes and halocarbons) concentrations (Felton, 1995; Lee et al., 2001; Zhao et al., 2007a; Guo et al., 2004).

In the study of Lee et al. (2001) Total Hydrocarbon (THC), benzene, toluene, chloroform, methylene chloride indoor concentration levels were found in four different styles of restaurants. In a Korean barbeque restaurant where cooking method of food was frying, mean concentrations of the target pollutants were found as $11.4 \,\mu\text{g/m}^3$, $18.4 \,\mu\text{g/m}^3$, $156 \,\mu\text{g/m}^3$, $14.9 \,\mu\text{g/m}^3$, and $0.6 \,\mu\text{g/m}^3$ during frying, respectively. Except for methylene chloride the VOC concentrations were found to be higher than those measured in the other restaurants. In our study, toluene, benzene, chloroform,

methylene chloride concentrations were determined in the first campaign of the study as $4.37~\mu g/m^3$, $3.88~\mu g/m^3$, $0.75~\mu g/m^3$, $1.83~\mu g/m^3$ and in the second campaign of the study as $3.59~\mu g/m^3$, $1.58~\mu g/m^3$, $0.21~\mu g/m^3$, $0.03~\mu g/m^3$, respectively. Only methylene chloride concentration that determined in the first campaign of this study is higher than methylene chloride concentration reported by Lee et al. (2001).

The measured TVOC levels are compared with the guideline values. In Table 5.1. International indoor air TVOC standards or indoor air quality guideline values and international occupational health standards are shown (Health Canada, 1995; Hong Kong, 2003; Charles et al., 2005). The mean TVOC level that was determined as 207 μg/m³ in the first campaign of the study is higher than the limit value of the excellent class value which was determined by Hong Kong Environmental Protection Department (HKEPD). In Table 5.2. International Indoor Air VOC Standards and Occupational Health Standards are shown for individual compounds. All determined VOC concentrations in this study are lower than the international indoor air limit values. The TVOC values determined in this study are in the first group which is the comfort class. However, it should be noted that the indoor air standards are 8-hr average values. Our concentrations determined by the sampling method are one hour averages, therefore, they are not directly comparable. Molhave et al., (1997) suggested the following TVOC classification of four groups. $C_{TVOC} \leq 200~\mu\text{g/m}^3$ as comfort class; $C_{TVOC} = 200\text{--}3000$ $\mu g/m^3$ as irritation and discomfort if other factors exist; $C_{TVOC} > 3000-25000 \ \mu g/m^3$ as discomfort range, and $C_{TVOC} > 25000 \ \mu\text{g/m}^3$ as toxic range.

Table 5.1. International Indoor Air TVOC Standards and Occupational Health Standards

Standards					
Indoor air Quality Parameter	Mean Concentrations (μg/m³)	Mean Concentrations (ppb)	Limit values	Average Time (Hour)	Reference
TVOC	Campaign-1	Campaign-1	87 / 261 ^a (ppb)	8	Hong Kong
TVOC	207	11.0-56.7	$200/600^{a}$ (µg/m ³)	8	Hong Kong
	Campaign-2	Campaign-2	$300 (\mu g/m^3)$	8	EC
	155	7.69-9.60	$600 / 3000^{b}$ (µg/m ³)	8	HKEPD

a) Guideline value for Excellent Class / Guideline value for Good Class of IAQ (Hong Kong, 2003)

b) Level I / Level II (HKEPD)

⁽**Hong Kong**): The Government of the Hong Kong Special Administrative Region, (2003), (**EC**): The European Comminity, (**HKEPD**): The Hong Kong Environmental Protection Department, (1999)

Table 5.2. International Indoor Air VOC Standards and Occupational Health Standards

Indoor air Quality Parameter (μg/m³)	ACGIH (8-hr)	OSHA (8-hr)	OEHHA (8-hr)	WHO	Japan (8-hr)	Hong Kong (8-hr)
benzene	2000	32000	60	16.1	16.1	-
toluene	189000	754000	300	260 [1 wk]	260	1902
n-butanol	61000	300000	-	-	-	-
n-butyl acetate	71000	710000	-	1	-	
chloroform	49000	240000	300	1	-	163
ethyl benzene	434000	435000	2000	22000 [1 yr]	3800	1447
ethyl acetate	1442000	1400000	-	-	-	
n-Heptane	1640000	2000000	-	1	-	-
n-Hexane	176000	1800000	7000	ı	-	-
isooctane	1402000	-	-	ı	-	-
nonane - all isomers	1050000	-	-	ı	-	-
n-Octane	1402000	2350000	-	-	-	-
alpha-Pinene	112000	-	-	-	-	-
beta-Pinene	112000	_	-	-	-	-
2-propanol	492000	980000	7000	-	-	-
o/m/p-xylene	434000	-	-	-	-	-

(ACGIH): The American Council of Governmental Industrial Hygienists, (ACGIH, 2004), (OSHA): The U.S. Occupational Health and Safety Administration, (OSHA, 2004), (OEHHA): Office of Environmental Health Hazard Assessment, (OEHHA, 2005), (WHO): World Health Organization Guidelines for Air Quality, (WHO, 1999), (Japan): Ministry of Health, Labour and Welfare of Japan, (MHLW, 2004), (Hong Kong): The Government of the Hong Kong Special Administrative Region, (2003)

5.2. Aldehyde Concentrations

The 15 target aldehydes were formaldehyde, acetaldehyde, acrolein, acetone, propionaldehyde, crotonaldehyde, butryaldehyde, benzaldehyde, isovaleraldehyde, valeraldehyde, o-tolualdehyde, m/p-tolualdehyde, hexaldehyde, and 2.5dimethylbenzaldehyde. Fourteen aldehydes were detected except for acrolein in the restaurant kitchen in all periods of sampling. Aldehydes that had the highest increase in concentration and also the highest concentrations in the during frying sampling period in the 1st campaign were acetaldehyde, formaldehyde, and hexaldehyde with average concentrations of 13.1, 2.95, and 1.29 µg/m³, respectively. In the second campaign of the study, aldehydes with the highest increase in concentrations and the highest concentrations in the during frying period were hexaldehyde, acetaldehyde, valeraldehyde, butryaldehyde, benzaldehyde with average concentrations were 0.86, 0.85, 0.32, 0.26, 0.15, and $0.13 \mu g/m^3$, respectively. Three of the fifteen target aldehydes (acrolein, crotonaldehyde and isovaleraldehyde) were not detected in all periods of sampling in the second campaign of the study. In addition, total aldehydes were determined as the sum of detected target aldehyde compounds as $19.4 \,\mu g/m^3$ and $4.42 \,\mu g/m^3$ in the first campaign and in the second campaign of the study in the during frying period, respectively.

Svendsen et al. (2002) determined aldehyde concentrations in nineteen restaurant kitchens where the french fries were fried in deep fryers with soy oil, sunflower oil, and palm oil. The highest aldehyde concentrations were determined at the breathing zone of the cook in a small local restaurant kitchen that was serving mostly fried food and had a insufficient ventilation system. The mean concentrations of formaldehyde, acetaldehyde and total aldehydes were found as $14 \,\mu\text{g/m}^3$, $102 \,\mu\text{g/m}^3$ and $119 \,\mu\text{g/m}^3$, respectively. In the two campaigns of this study formaldehyde, acetaldehyde and total aldehyde concentration levels were lower than the aldehyde concentrations determined by Svendsen et al. (2002). Lee et al. (2001) determined average formaldehyde concentration as $177 \,\mu\text{g/m}^3$ during frying in a Korean barbeque restaurant where the food was fried in a pan. This formaldehyde concentration is higher than that measured in this study.

The final report of National Research Council Canada (Charles et al., 2005) summarized some of the most well-established guidelines and standards relating to indoor air quality. In this report International indoor air aldehyde standards and occupational health standards were presented which are shown in Table 5.3. According to Office of Environmental Health Hazard Assessment (OEHHA) of California EPA limit values, acetaldehyde indoor air concentration must be below 9 μ g/m³. In the first campaign of this study, acetaldehyde concentration during frying was found as 13.1 μ g/m³ which is higher than acetaldehyde limit value of OEHHA.

Table 5.3. International Indoor Air Aldehyde Standards and Occupational Health Standards

Indoor air Quality Parameter (µg/m³)	ACGIH (8-hr)	OSHA (8-hr)	OEHHA (8-hr)	WHO	Japan (8-hr)
acetaldehyde	45000	360000	9	50 [1 yr]	48
formaldehyde	400	900	30	100 [30 min]	100
acetone	1188000	2400000	-	-	-
propionaldehyde	48000	-	-	-	-
valeraldehyde	176000	ı	-	-	-
crotonaldehyde	900	6000	-	-	-

(ACGIH): The American Council of Governmental Industrial Hygienists, (ACGIH, 2004), (OSHA): The U.S. Occupational Health and Safety Administration, (OSHA, 2004), (OEHHA): Office of Environmental Health Hazard Assessment, (OEHHA, 2005), (WHO): World Health Organization Guidelines for Air Quality, (WHO, 1999), (Japan): Ministry of Health, Labour and Welfare of Japan, (MHLW, 2004)

5.3. PM Concentrations

In this study, we measured particles that have aerodynamic sizes of <2.5μm which is considered as respirable and <10μm considered as inhalable for human beings. In the first campaign of the study in real life conditions; frying of french fries, hamburger patties, chicken patties, schnitzels and nuggets with the vegetable margarine in a deep fryer produced high concentrations of particulate matter in both of the size fractions (PM_{2.5} and PM₁₀) which were shown in Table 4.6. and in Table 4.7 in Section 4. In the second campaign of the study only french cut potatoes were fried with varying amounts (1250g, 2500g and 3750g). The measured PM_{2.5} and PM₁₀ levels were presented in Table 4.18. and Table 4.19. According to the results we can say that concentration levels of PM_{2.5} and PM₁₀ directly proportional to the amount of french fries that were fried. When we compare first campaign PM_{2.5} and PM₁₀ concentrations with the second campaign of the study, higher concentrations were measured in the second campaign. These results show that the amount of fried food is an important factor for indoor particulate matter concentrations.

Others (Ozkaynak et al., 1996a,b; Brauer et al., 2000; Kelly, 2001; Lee et al., 2001; Fortmann et al., 2002; Long et al.; 2000; Wallace et al., 2004; Evans et al., 2008) have studied levels of particulate matter in the breathing zone of the cook or indoor concentrations in kitchen during frying activity. The study that was performed by Lee et al., (2001) in different styles of restaurants found the highest particulate matter concentration levels in a Korean barbeque restaurant where the food was fried in a pan.

The highest indoor levels of PM₁₀ and PM_{2.5} were found as 1442 μ g/m³ and 1167 μ g/m³, respectively. In two campaigns of our study determined PM₁₀ concentration levels were found to be higher than those measured by Lee et al. (2001) but PM_{2.5} concentration levels were lower. Another study was performed by Kelly (2001) with frying hamburgers on a gas stove and a electric stove which produced average PM_{2.5} concentrations of 115 μ g/m³ and 230 μ g/m³, and PM₁₀ concentrations of 118 μ g/m³ and 328 µg/m³, respectively in a research house kitchen at the breathing zone of the cook. In this study the highest average level of $PM_{2.5}$ concentration was found as 249 $\mu g/m^3$ which is higher than the Kelly (2001) study results. Fortmann et al. (2002) performed their study in a test house. One of the sampling locations was kitchen air and the second one was at the breathing zone of the cook during frying of french fries with vegetable oil in a deep fryer on a electric or gas stove. PM_{2.5} concentrations at the breathing zone of the cook were four to six times higher than the kitchen air concentrations however PM₁₀ indoor air concentrations were higher than the determined PM₁₀ concentrations at breathing zone of the cook. The average $PM_{2.5}$ concentrations were found as 374 $\mu g/m^3$, $66 \mu g/m^3$, $123 \mu g/m^3$ and PM_{10} concentrations were found as $207 \mu g/m^3$, $724 \mu g/m^3$, and 715 $\mu g/m^3$ at the breathing zone of the cook during frying, in the kitchen air during frying, and average of total exposure period in the uses of electric stove respectively. Average concentrations of PM_{2.5} in this study were lower, however PM₁₀ concentrations were higher than the concentrations reported by Fortmann et al. (2002).

Lastly, we compared concentrations measured in this study with the international indoor air quality standards or indoor air quality guideline values and international occupational health standards. The comparison is shown in Table 5.4. (OSHA, 2004; Charles et al., 2005) and Table 5.5. (Health Canada, 1995; Hong Kong, 2003; Charles et al., 2005). It can be said that indoor air PM_{2.5} concentration levels were found to be lower than the occupational health limits that were determined by Occupational Safety and Health Administration (OSHA) and The American Council of Governmental Industrial Hygienists (ACGIH), but they were found to be higher than indoor air quality limit values. PM₁₀ concentration levels measured in this study were higher than most of the indoor air quality limit values except for (ACGIH) limit values. However, the limits are 8-hr average values while PM₁₀ measured in this study is 4-hr average concentrations.

Table 5.4. International Indoor Air PM_{2.5} Standards and Occupational Health Standards

Indoor air Quality Parameter	Mean Concentrations (μg/m³)	Limit values (µg/m³)	Average Time (hour)	Reference
PM _{2.5}	Campaign-1	5000	8	OSHA
	108	3000	8	ACGIH
	Campaign-2	100	1	НС
	147	65	24	EPA

(OSHA): The U.S. Occupational Health and Safety Administration, (ACGIH): The American Council of Governmental Industrial Hygienists (2001), (HC): Health Canadai, (1995), (EPA): Environmental Protection Agency (NAAQS)

Table 5.5. International Indoor Air PM₁₀ Standards and Occupational Health Standards

Indoor air Quality Parameter	Mean Concentrations (μg/m³)	Limit values (μg/m³)	Average Time (Hour)	Reference
PM ₁₀	Campaign-1	150	24	EPA
	1583	100	1	Health Canada
	Campaign-2	20/180 a	8	Hong Kong
	5778	10000	8	ACGIH

a) Guideline value for Excellent Class / Guideline value for Good Class of IAQ (Hong Kong, 2003)
(EPA): Environmental Protection Agency (NAAQS), (HC): Health Canada (1995), (Hong Kong): The Government of the Hong Kong Special Administrative Region (2003), (ACGIH): The American Council of Governmental Industrial Hygienists, (2001)

5.4. Inorganic Gas Concentrations

In the first campaign of the study, the mean CO₂ concentrations were measured in the range from 532 ppm to 559 ppm, while in the second campaign of the study, mean CO₂ concentrations were ranged from 423 ppm to 487 ppm. Mean CO concentrations were ranged from 2.12 ppm and 2.67 ppm and from 0.08 ppm to 0.33 ppm in during frying periods of the first campaign and the second campaign of the study respectively.

There are some studies that measured CO and CO₂ indoor air concentrations in restaurant or house kitchens during frying activity (Kelly, 2001; Fortmann et al. 2001; Lee et al., 2001). CO concentrations were determined as inorganic pollutants in restaurants when during various types of food (french fries, bacon, tortillas, fish, beef, pork roast and popcorn) were cooked in the study by Fortmann et al. (2001). Mean CO

concentrations were measured as 1 ppm and 0.8 ppm during frying of french fries and total exposure level, respectively.

When we look at the international indoor air CO₂ standards or indoor air quality guideline values and international occupational health standards which are shown in Table 5.6, the mean CO₂ concentration in during frying study in all campaigns of the study are lower than the international indoor air limit values. (Charles, et al. 2005, Hong Kong 2003, OSHA 2004).

International indoor air CO standards or indoor air quality guideline values and international occupational health standards are shown in Table 5.7 (Charles et al., 2005; Health Canada, 1995; WHO, 2000). Indoor air limit value that was determined by the Government of the Hong Kong Special Administrative Region (Hong Kong) as 1.7 ppm for excellent class and by German Federal Environmental Agency (German) as 1.3 ppm which was guideline value I (concentration of a substance in indoor air for which, when considered individually, there is currently no evidence that even life-long exposure is expected to cause any adverse health impacts). Determined mean CO concentration in the first campaign of the study is higher than the limt indoor air CO values that were determined by German and Hong Kong autohorities.

Table 5.6. International Indoor Air CO₂ Standards and Occupational Health Standards

Indoor air Quality Parameter	Mean Concentration Range (ppm)	Limit values (µg/m³)	Average Time (hour)	Reference
	Campaign-1	800/1000 ^a	8	Hong Kong
CO_2	532-559	5000	8	OSHA, NIOSH, ACGIH
	Campaign-2			
	423-487	10000	1	DFG

a) Guideline value for Excellent Class / Guideline value for Good Class of IAQ (Hong Kong, 2003) b) Level I / Level II (HKEPD)

⁽Hong Kong): The Government of the Hong Kong Special Administrative Region, (2003), (EC): The European Comminity, (HKEPD): The Hong Kong Environmental Protection Department, (1999), (OSHA): The U.S. Occupational Health and Safety Administration, (NIOSH): U.S. National Institute for Occupational Safety and Health, (1992), (ACGIH): The American Council of Governmental Industrial Hygienists, (2001), (DFG): Deutsche Forschungs Gemeinschaft, (2000)

Table 5.7. International Indoor Air CO Standards and Occupational Health Standards

Indoor air Quality Parameter	Mean Concentration Range (ppm)	Limit values (ppm)	Average Time (Hour)	Reference
СО	Campaign-1	25	1	HC
		35	1	EPA
	2.12-2.67	25	1	WHO
		52/5.2 ^b	0.5	German
	Campaign-2	11	8	HC
		30	8	DFG
	0.08-0.33	1.7/8.7 ^a	8	Hong Kong
		25	8	ACGIH

a) Guideline value for Excellent Class / Guideline value for Good Class of IAQ (Hong Kong, 2003)

b) Guideline value II / Guideline value I, (German, 2005)

⁽EPA): Environmental Protection Agency, (NAAQS), (DFG): Deutsche Forschungs Gemeinschaft , (2000), (HC): Health Canada, (1995), (WHO Europe): The World Health Organization , (2000), (Hong Kong): The Government of the Hong Kong Special Administrative Region, (2003), (German): German Federal Environmental Agency, (2005), (ACGIH): The American Council of Governmental Industrial Hygienists, (2001)

CHAPTER 6

CONCLUSIONS

Volatile Organic Compound (VOC), aldehyde, and PM_{2.5} concentrations were determined with active sampling; TVOC, CO₂, and CO concentrations, and temperature and relative humidity levels were monitored with a continuous air monitoring device in a restaurant kitchen, in two campaigns. Campaign-1 was conducted to characterize the cook's exposure concentrations. Campaign-2 was conducted under controlled conditions: only French cut potatoes were fried at daily varying amounts (1st day, 1250 g; 2nd day, 2500 g; and 3rd day, 3750 g).

The VOCs detected with the highest mean concentrations in the kitchen were nheptane, ethyl acetate, nonanal, n-octane, toluene, decanal, and n-decane during frying in the first campaign with the mean concentrations of 83.0, 24.9, 23.4, 16.6, 4.37, 3.12, and 1.91 μg/m³, respectively. In the second campaign, the VOCs with the highest mean concentrations during frying were n-heptane, nonanal, n-octane, ethyl acetate, limonene, n-butanol, and n-undecane with the mean concentrations of 57.25, 19.4, 12.38, 3.54, 1.57, 1.27, and 1.13 μg/m³, respectively. Four of the seven VOCs, which are n-heptane, nonanal, n-octane, ethyl acetate, were the common compounds among the most increased ones of the two campaigns of the study. The difference in the concentrations between the two campaign may be due to the difference in the fried food. Frying of hamburger patties, chicken patties, nuggets, and schnitzels in Campaign-1 could have resulted in higher n-heptane, nonanal, n-octane, and ethylacetate concentrations.

Aldehydes that had the highest increase in concentration from before to during frying periods were acetaldehyde, formaldehyde and hexaldehyde, in the first campaign, respectively. Their concentrations during frying were 13.1, 2.95 and 1.29 μg/m³, respectively. In Campaign-2, aldehydes with the highest increase in concentrations in during frying period were hexaldehyde, acetaldehyde, valeraldehyde, butryaldehyde, benzaldehyde, and 2,5-dimethylbenzaldehyde with the concentrations of 0.86, 0.85, 0.32, 0.26, 0.15, 0.13 μg/m³, respectively. In addition, Total VOCs (TVOCs) and total aldehydes concentrations were calculated. TVOCs mean concentration was determined in the first campaing as 139 μg/m³, 207 μg/m³, and 144 μg/m³, and in the second

campaign as 140 $\mu g/m^3$, 155 $\mu g/m^3$, and 106 $\mu g/m^3$ for before frying, during frying and after frying sampling periods, respectively. Total aldehydes concentration was determined as 19.4 $\mu g/m^3$ and 4.42 $\mu g/m^3$ in during frying period in the first campaign and in the second campaign, respectively. These levels are not very different from levels reported in the literature for other indoor environments, therefore can be considered as not significantly related to cooks' exposure due to frying.

 $PM_{2.5}$ concentrations were determined by using 4-hr samples collected with an active sampler. The mean $PM_{2.5}$ concentration of three days sampling was $108~\mu g/m^3$ and median $PM_{2.5}$ concentration was $90.0~\mu g/m^3$ in Campaign-1. The mean three day PM_{10} concentrations in the three monitoring periods was ranged from 694 to 1583 $\mu g/m^3$ in the first campaign. In the second campaign PM_{10} concentrations were ranged from 724 $\mu g/m^3$ to 3292 $\mu g/m^3$, from 324 $\mu g/m^3$ to 5554 $\mu g/m^3$, from 1814 $\mu g/m^3$ 9070 $\mu g/m^3$ in the 1st, 2nd, 3rd days of the campaign in which for 1250 g, 2500 g and 3750 g french cut potatoes were fried, respectively. In this campaign, $PM_{2.5}$ concentrations were determined as 81.3 $\mu g/m^3$, 110 $\mu g/m^3$, 249 $\mu g/m^3$ in the first, the second and the third day, respectively. The mean of the three day $PM_{2.5}$ concentration was determined as 147 $\mu g/m^3$. The increase in the PM concentrations indicate that emission of particles is related to the amount of potatoes fried, and the realtively high levels of PM indicate that cooks' exposure during frying may result in chronic health effects of pollutant emissions occur during frying, further research is needed in order to protect the cooks from consequences.

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