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Speciated non-methane organic compounds emissions from food cooking in Mexico

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Abstract

Non-methane organic compound (NMOC) emissions from different sorts of food preparation sites, were quantified for the first time in Mexico, in order to develop emission profiles for further application in the chemical mass balance receptor model (CMB). Restaurants using charcoal grills and LP gas stoves, “tortillerías”, food frying places and rotisseries were sampled using SUMMA[®] stainless-steel canisters to analyse NMOC by high-resolution gas chromatography. The results obtained show that profiles determined from food cooking processes have similarities to those found in LP gas combustion, which is the most common fuel in Mexico used for this purpose, although there were differences in the relative composition of propane and butane in both cases. This suggests that, the rates of combustion of propane and butane are different. It has also been detected that propene, a reactive olefin is produced during the combustion process. The obtained profiles of restaurants, rotisseries and fried food show an important contribution of two carbon compounds (ethane, ethylene and acetylene) that can be attributed to the complex process of grease and meat cooking. The presence of these compounds cannot be attributed to vehicular sources since the concentrations are higher than in ambient air. These were also determined from aromatic compounds such as benzene, toluene and xylene in the combustion of vegetal charcoal. The measured concentrations indicate that NMOC emissions from cooking may become an important indoor source of NMOC under crowded conditions in closed places. © 2001 Published by Elsevier Science Ltd.

Keywords: NMOC emission; Air pollution; Mexico City; Source profiles

1. Introduction

In addition to pollutants emitted from several sources there exist many photochemical reactions that produce ozone and other photochemical oxidants that add to an already complex urban air pollution problem. The non-methane organic compounds (NMOC) play an important role in this photochemical smog formation (Seinfeld and Pandis, 1997). Airshed dispersion models and chemical mass balance receptor model (CMB) (Watson et al.,

1991; Fujita et al., 1994) have been developed and used to understand some of the relationships between the pollutants emitted by different sources and the secondary aerosols formed as a result of photochemical reactions. One of the limiting factors in the application of these types of models, in particular, receptor models is that a unique NMOC emission profile must be derived for each source and this information is very scarce in Mexico.

Mexico City is considered the most polluted urban area in the world, where the standard of ozone is exceeded more than 300 days every year (DDF, Proaire, 1997). Some of the most important sources of NMOC, such as gasoline-powered motor vehicles, LP gas, dry cleaning, solvent use, asphalt operations, and others, have been characterised to understand the origin and

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variation of these compounds in urban areas and also for modelling purposes (Mugica et al., 1998 a, b; Vega et al., 2000), but there is no information about other sources such as of food cooking, while due to the large amount of sites involved, it also becomes an important NMOC source. In Mexico City there are more than 30,000 registered places where food is cooked and more than 2500 small informal places that prepare all kinds of meals on the sidewalks of the City. Frequently, large smoke plumes coming from these sites can be observed, although some restaurants have chimneys to avoid disturbances to the consumers, there are no control mechanisms to reduce the emissions from these sources to the atmosphere.

In Mexico, there is no information about the NMOC emissions from food preparation and in other countries such as the United States very limited data exists concerning these emissions: some of them being the studies on emissions from hamburgers cooking and food-frying operations (Schauer et al., 1998) and the Northern Front Air Quality Study carried out in winter 1997. In this study gaseous organic compounds were characterised to construct specific source profiles used in receptor modelling (Zielinska and McDonald, 1999). As there are so many differences in the kind of food, oils and fuels used in Mexico, it would be imprecise to use the information generated in the US for Mexico City CMB modelling. Other authors have researched about the health risks that the NMOC emissions from stoves could result (Zhang and Smith, 1996).

Therefore, the main objective of the present work is to determine the chemical composition of the NMOC emitted during cooking operations to obtain the emission profiles which could be used for CMB receptor model application. This project provides the first data on chemical species of NMOC from cooking places and could be useful also in designing emission control equipment in these commercial facilities.

2. Methods

A variety of food preparation sites in Mexico City was selected to obtain representative NMOC emission profiles: two tortillerias where LP gas is used, two restaurants using both, LP gas stove and charcoal grill, which cooked many popular Mexican dishes; one chicken roasting place using LP gas as fuel, and finally, one site where pork lard was used for frying meat and Mexican snacks. Samples were taken during the period from 20–31 March in 1998.

NMOC sampling and analysis were carried out according to the TO-14 USEPA protocol (Winberry et al., 1988). Samples for emission-profile characterisation were collected directly at the sites of the sources, using 61 SUMMA® canisters. The collection of ambient air samples was conducted using programmable AN-

DERSEN equipment, with a flow regulator of critical orifice that allows adjustment of the incoming flow in the range of 2–500 ml min⁻¹. This facilitated the control over the sample time, which in turn was regulated in accordance with the overall activity of the site elected. Emissions were sampled downstream from the extractor in the exhaust ventilation system using a Teflon pipe. Prior to testing, pipes and connections were cleaned carefully and canisters were cleaned by several cycles of vacuum with ENTECH equipment.

Samples were analysed in the Mexican Petroleum Institute (IMP), using a Hewlett-Packard Gas Chromatograph (5890 Series Plus II) after concentration by collection in a freeze-out loop made from chromatographic grade stainless-steel tubing packed with 60/80 mesh deactivated glass beads. The cryogen was removed and the NMOC collected were revolatilised and separated on a GC column equipped with a flame ionisation detector (FID). The chromatographic capillary column used for C₂–C₁₂ hydrocarbon analysis was a J&W DB-1, made of fused silica, with an internal diameter of 0.32 mm, length of 60 m, and 1 µm phase thickness. The oven temperature program was –50°C for 3 min, to 200°C at 8°C min⁻¹. A separate gas chromatograph (TREMTRICS, Series 9000 with a chrompack Plot fused silica capillary column of 60 m length and an internal diameter of 0.32 mm) was used to obtain accurate concentrations for C₂ hydrocarbons. A single response factor was determined for the column and was used to calculate NMOC component concentration as parts per billion of carbon (ppb C). This has been demonstrated to be valid for NMOC compounds (Lonneman et al., 1986). The chromatograph was calibrated by injecting known amounts of certified standard calibration gases from the US Environmental Protection Agency (US EPA) in the chromatography column.

3. Sites description

3.1. Tortillerias

Two tortillerias with LP gas stoves with transport bands were selected. Inside the tortillerias there was a machine, which was used to process corn meal dough through a rolling mill that produced tortillas continuously which were accommodated over a metal band that circulates through a heated oven like vestibule. After this, the tortillas were sent to collecting trays to allow their weighing before the customer. The first tortilleria had two stoves and its own mill in an area of 30 m². During the 3.3 h of sampling the work was continuous. The second one had only one stove and 10-m² area. During the 3 h-sampling period the band was stopped several times because there were few customers.

3.2. Restaurants

Four samplings were carried out in two restaurants in Mexico City. These restaurants had both LP gas stoves and charcoal grill for cooking meat and other foodstuffs. The first one had an area of 80 m² and 20 tables, the sampling periods were 60 min. The second restaurant had an area of 50 m² and the first sampling period was 1 h and the second one was 2 h. In both the restaurants there were stacks on the roof over the grill.

3.3. Rotisserie

The place had two roasting ovens in an area of 10 m². There were four sampling periods of 1 h each. The samples were taken near the exit of the exhaust system. The campaign was done on Sunday and there was no traffic around the site.

3.4. Fried food

Five 1-h integrated samples were taken in an area of 10 m², where food-frying operations employing pork lard were carried out. The food consisted mainly of pork meat, pork skin (known as chicharron) and some fries. There was no ventilation system and the pipe was located at 2-m height over the cooking pot. The place was close to an avenue.

4. Results and discussion

More than 100 compounds were quantified, but in this paper only the most important ones are reported because they represent more than 80% of the total mass. There are some unknown compounds especially in restaurants and rotisserie, and other compounds are in very small concentrations. Levels of the background were con-

sidered negligible because the samples were taken from the exhaust of the chimney of the different places. Table 1 shows the average chemical composition of the source profiles of food cooking. The components (propane, butane and isobutane) of the LP gas were the most abundant species in all the cases. Propene was present in abundance seven times higher than in the raw fuel. In general, it can be observed that there is an important contribution of two carbon compounds, except in tortillerias. The largest amount of these was measured at the rotisseries with 28%, followed by restaurants and fried food places with 17%. Although the sites were closer to avenues, there was low traffic and the sampling was carried out very close to the exhaust systems or over the pot in the case of fried pork, therefore interference's were negligible. This means that the compounds reported came from the food and/or fuel used during the processes. The results suggest that tortillerias' profile appears to resemble the result of combustion of LP gas and there is no NMOC contribution from the tortillas. This is believed because there is little contribution of two carbon compounds in this source (only due to the fuel combustion) in comparison with the larger contribution of these compounds in rotisseries and food fried places that use the same fuel. What rotisseries, restaurants and fried food have in common is that cooking of meals is more complex, there is a lot of grease involved during the process and these could be the possible source of increase of the two carbon compounds. A comparison of some emitted species is shown in Fig. 1.

The tortilleria profile showed higher propane, *n*-butane and *iso*-butane content of 53, 24.4 and 9.15%, respectively. These results are associated with the LP gas profile, although during the fuel combustion process the proportion between the compounds changed from the original profile. This means that the combustion rate of these compounds is not the same. The contribution of two carbon compounds was very low, 1% in one

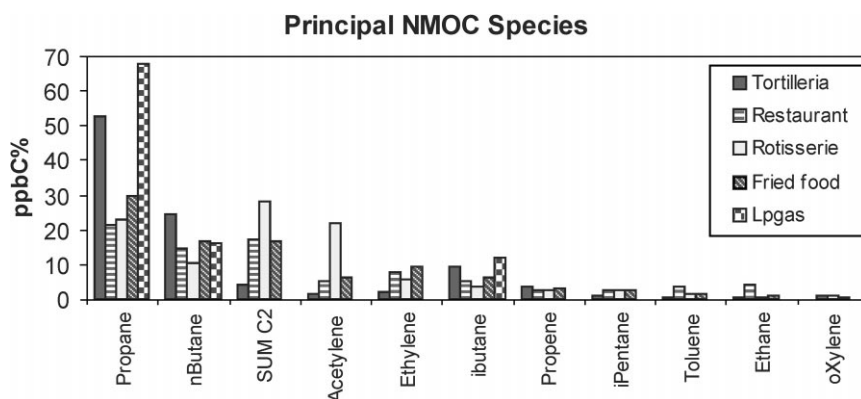


Fig. 1. Species emitted from different cooking place.

Table 1
NMOC emission profiles for food cooking (ppb C%)

Species	Restaurant average	Tortilleria average	Rotisserie average	Fried food average	LP Gas profile
Ethane	4.11 ± 2.69	0.34 ± 0.47	0.77 ± 0.16	1.24 ± 0.25	0.98 ± 1.12
Ethylene	8.04 ± 3.36	2.00 ± 2.70	5.61 ± 1.82	9.20 ± 3.16	0.00 ± 0.00
Acetylene	5.21 ± 2.41	1.66 ± 2.12	21.85 ± 13.14	6.37 ± 3.46	0.00 ± 0.00
C2 sum	17.3 ± 5.80	3.99 ± 1.29	28.23 ± 14.65	16.80 ± 5.86	0.98 ± 1.12
Propene	2.77 ± 0.98	3.81 ± 3.38	2.55 ± 0.29	2.92 ± 0.82	0.43 ± 0.29
Propane	21.45 ± 10.63	52.84 ± 0.28	22.89 ± 8.26	30.00 ± 4.27	67.66 ± 7.70
<i>i</i> -Butane	5.0 ± 2.79	9.15 ± 3.23	3.52 ± 1.19	6.23 ± 0.83	12.07 ± 3.75
<i>n</i> -Butane	14.4 ± 7.60	24.42 ± 9.14	10.25 ± 3.46	16.62 ± 1.86	16.30 ± 4.04
<i>trans</i> -2-Butene	0.20 ± 0.08	0.17 ± 0.00	0.23 ± 0.07	0.11 ± 0.07	0.81 ± 0.44
<i>cis</i> -2-Butene	0.13 ± 0.06	0.10 ± 0.00	0.15 ± 0.03	0.10 ± 0.06	0.48 ± 0.25
<i>I</i> -Pentane	2.38 ± 0.89	1.06 ± 0.00	2.45 ± 0.25	2.85 ± 0.24	0.14 ± 0.11
1-Pentene	0.17 ± 0.20	0.01 ± 0.01	0.07 ± 0.02	0.07 ± 0.05	0.00 ± 0.00
2-Methyl-1-butene	0.14 ± 0.10	0.03 ± 0.03	0.13 ± 0.11	0.14 ± 0.16	0.00 ± 0.00
<i>n</i> -Pentane	0.97 ± 0.56	0.23 ± 0.00	1.02 ± 0.42	2.93 ± 1.17	0.02 ± 0.03
Isoprene	0.00 ± 0.00	0.00 ± 0.00	0.00 ± 0.00	0.00 ± 0.00	0.00 ± 0.00
2-Methyl-2-butene	0.13 ± 0.09	0.01 ± 0.01	0.13 ± 0.02	0.14 ± 0.05	0.00 ± 0.00
MTBE	0.56 ± 0.36	0.10 ± 0.11	0.89 ± 0.28	0.94 ± 0.30	0.00 ± 0.00
Cyclopentene	0.30 ± 0.23	0.02 ± 0.02	0.21 ± 0.15	0.17 ± 0.10	0.00 ± 0.00
2-Methyl-1-pentene	0.41 ± 0.34	0.01 ± 0.00	0.08 ± 0.10	0.05 ± 0.04	0.00 ± 0.00
Cyclopentane	0.00 ± 0.00	0.00 ± 0.00	0.00 ± 0.00	0.00 ± 0.00	0.00 ± 0.00
2-Methylpentane	0.47 ± 0.31	0.08 ± 0.08	0.62 ± 0.15	0.67 ± 0.26	0.00 ± 0.00
3-Methylpentane	0.40 ± 0.30	0.05 ± 0.05	0.41 ± 0.08	0.38 ± 0.14	0.00 ± 0.00
<i>n</i> -Hexane	0.94 ± 0.69	0.09 ± 0.09	0.80 ± 0.08	0.85 ± 0.16	0.00 ± 0.00
<i>trans</i> -2-Hexene	0.01 ± 0.02	0.00 ± 0.00	0.00 ± 0.00	0.01 ± 0.01	0.00 ± 0.00
Methylcyclopentane	0.01 ± 0.02	0.02 ± 0.02	0.05 ± 0.06	0.01 ± 0.02	0.00 ± 0.00
2,4-Dimethylpentane	0.06 ± 0.06	0.01 ± 0.01	0.15 ± 0.05	0.14 ± 0.06	0.00 ± 0.00
Benzene	3.19 ± 2.22	0.26 ± 0.34	1.01 ± 0.19	0.70 ± 0.29	0.00 ± 0.00
2-Methylhexane	0.14 ± 0.07	0.03 ± 0.03	0.27 ± 0.02	0.21 ± 0.09	0.00 ± 0.00
2,3-Dimethylpentane	0.09 ± 0.04	0.02 ± 0.02	0.19 ± 0.06	0.17 ± 0.07	0.00 ± 0.00
3-Methylhexane	0.25 ± 0.11	0.04 ± 0.04	0.68 ± 0.68	0.44 ± 0.25	0.00 ± 0.00
2,2,4-Trimethylpentane	0.44 ± 0.28	0.08 ± 0.09	0.76 ± 0.29	0.81 ± 0.41	0.00 ± 0.00
<i>n</i> -Heptane	0.33 ± 0.23	0.03 ± 0.03	0.53 ± 0.22	1.40 ± 0.69	0.00 ± 0.00
2,3,4-Trimethylpentane	0.1 ± 0.09	0.03 ± 0.03	0.32 ± 0.13	0.29 ± 0.15	0.00 ± 0.00
Toluene	3.88 ± 2.91	0.31 ± 0.35	1.69 ± 0.24	1.76 ± 0.53	0.00 ± 0.00
3-Methylheptane	0.02 ± 0.02	0.01 ± 0.01	0.19 ± 0.16	0.01 ± 0.03	0.00 ± 0.00
<i>n</i> -Octane	0.24 ± 0.16	0.01 ± 0.01	0.58 ± 0.33	1.34 ± 0.75	0.00 ± 0.00
Ethylbenzene	0.73 ± 0.61	0.05 ± 0.05	0.57 ± 0.09	0.51 ± 0.29	0.00 ± 0.00
Estirene	0.23 ± 0.17	0.01 ± 0.01	0.20 ± 0.12	0.08 ± 0.10	0.00 ± 0.00
Oxilene	1.09 ± 0.77	0.07 ± 0.06	0.81 ± 0.17	0.66 ± 0.36	0.00 ± 0.00
<i>n</i> -Nonane	0.12 ± 0.06	0.01 ± 0.01	0.39 ± 0.30	0.10 ± 0.07	0.00 ± 0.00
β -Pinene	0.37 ± 0.27	0.04 ± 0.04	0.58 ± 0.09	0.28 ± 0.09	0.00 ± 0.00
<i>n</i> -Propilbenzene	0.06 ± 0.06	0.01 ± 0.01	0.12 ± 0.03	0.06 ± 0.05	0.00 ± 0.00
1,3,5-TMBenzene	0.07 ± 0.07	0.01 ± 0.02	0.26 ± 0.11	0.06 ± 0.06	0.00 ± 0.00
Methyltoluene	0.18 ± 0.12	0.03 ± 0.03	0.35 ± 0.10	0.15 ± 0.05	0.00 ± 0.00
1,2,4-TMBenzene	0.22 ± 0.06	0.01 ± 0.01	0.22 ± 0.12	0.06 ± 0.05	0.00 ± 0.00
<i>n</i> -Decane	0.00 ± 0.01	0.00 ± 0.01	0.01 ± 0.03	0.01 ± 0.02	0.00 ± 0.00
<i>n</i> -Undecane	0.06 ± 0.06	0.01 ± 0.01	0.14 ± 0.04	0.05 ± 0.05	0.00 ± 0.00
Total ppb C% sum	80.07	97.28	84.7	91.29	98.89

tortilleria and 4% in a bigger one, maybe because the second one is next to a fried food place. In the bigger tortilleria the total concentration of NMOC rises to 260 ppm C.

As mentioned above, restaurant samples showed that there is a large contribution of two carbon compounds with 17% and although both restaurants were close to avenues, traffic was very low and samples were taken

very close to the exhaust systems. On the other hand, the concentrations for these compounds are higher than in the ambient air. The presence of ethane, ethylene and acetylene could be due to incomplete combustion of meals, grease and charcoal used in the restaurants. Propane contribution was 21%, *n*-butane was 14.4% and *iso*-butane 5%, these results are related to the LP gas profile, which is one of the most important fuels used in the restaurants (Valencia, 1997). It was found that there are some contributions of aromatic species, some due to vehicular sources, but others are due to the presence of these compounds in vegetal charcoal. This is believed because the detected concentrations of these compounds are slightly higher than in ambient air, and restaurants were the only sites that presented an important contribution of aromatic compounds. In this direction, other authors have reported the contribution of benzene to ambient air from wood combustion (Schauer et al., 1998). The total NMOC concentration found in restaurant samples was between 6 and 38 ppm C.

The roasted chicken place presented the most abundant two carbon compounds with 28% ppb C, especially acetylene. This abundance cannot be completely attributed to traffic because samples were collected on a Sunday and there was little traffic. In this place, the fuel used is LP gas and the presence of these compounds must be due to the incomplete combustion of meat and grease. The abundance of propane, *n*-butane and *iso*-butane were 23, 13 and 3.5%, respectively, and they are related to the LP gas profile. The average of total NMOC was 10.7 ppm C.

Finally, in the pork-meat-cooking place also there was a high abundance of two carbon compounds with 17% ppb C. This can be due, as in restaurant and rotisseries, to the incomplete combustion of grease. The abundance of propane, *n*-butane and *iso*-butane were 30, 17 and 6% related to the LP gas which is the fuel used. The total NMOC concentration of samples oscillated between 3 and 12 ppm C.

5. Conclusions

The NMOC composition of cooking emissions was measured in Mexico for the first time, in order to get more knowledge about these emissions, to obtain specific profiles of these processes and for further application of the CMB model. Different samples were taken in restaurants, tortillerias, rotisseries and fried food places to obtain the chemical composition of the emissions. Propane, *n*-butane and *iso*-butane, components of LP gas, were found to be the predominant compounds emitted from cooking operations, although with a relatively different composition of the species as referred to the original LP gas profile, indicating that the combustion rates of those

compounds are different. Propene was also found in higher abundance than in the original LP gas profile, which means that it is formed during the combustion of the fuel.

Ethylene, acetylene and ethane emissions were found in higher concentration than in ambient air for restaurants, rotisseries and fried food places, but not in tortillerias. This suggests that these compounds are produced from the combustion of food and grease. The presence of two carbon compounds in the ambient air has been always attributed to vehicle source, the results obtained in this study showed that the food cooking can be an important source of these compounds. Measurements also showed that when charcoal is used, there are some aromatic species emitted such as benzene, toluene and xylene, that cannot be attributed to vehicle sources.

The profiles obtained in this paper can be used with air quality measurements and profiles of other sources to determine the percentage contribution of food cooking emission applying the CMB model. These food-cooking profiles can be used in many localities of the country and any place with similar food preparation and fuels. It is believed that the cooking profiles at homes should be quite similar because traditional dishes prepared in the studied sites are the same as those prepared by thousands of families but it would be interesting to conduct a specific study about home stove emissions during cooking.

Although many of the cooking places have big open doors or windows, there are many restaurants and commercial food places where there are no ventilation or exhaust systems. This means that cooking operation may be an important indoor source of NMOC, and should be taken into account not only in ambient air quality studies but also in indoor air quality.

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