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Liquefied petroleum gas effect on ozone formation in Mexico city

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Abstract

An experimental study of outdoor smog chambers was carried out to determine effects of liquefied petroleum gas (LPG) on maximum ozone (O_3 max). 60% additions of commercial LPG and 60%-propane/40% butane mixture of the initial concentration were introduced into eight smog chambers containing morning ambient air of Mexico City metropolitan area (MCMA). The ozone concentrations in each chamber were monitored throughout the day to determine the maximum of ozone. Dilution experiments of 50% total hydrocarbons and associated compounds to LPG in morning ambient air were carried out too.

The results showed, that by increasing 60% of the associated compounds to LPG in the air of MCMA or diminishing 50% of them, had not an appreciable influence. The largest effect on ozone formation is determined by total nonmethane hydrocarbon (tNMHC) contained in the atmosphere, being the maximum of ozone formed in the smog chambers, on the average it diminished a 55%. C_3 and C_4 compounds associated to LPG only contribute a 14% of the total ozone formation.

Keywords: Smog chamber experiments; Air quality; Ozone precursors

1. Introduction

The ozone pollution problem in Mexico City has been associated with gasoline, due to exhaust and evaporative emissions, which include primary precursors, NO_x and NMHC in ozone formation. In 1993 a study was carried out in Mexico City by Blake and Rowland (1995), in which they concluded that C_3 and C_4 compound emissions associated with liquefied petroleum gas (LPG) leakage and incomplete combustion had a significant role, in causing the excessive ozone observed profiles. This led Petroleos Mexicanos-Gas (a subsidiary of the Mexican Petroleum Industry) to finance a project to search into the influence of LPG on ozone in Mexico City.

In the ambient air of Mexico City metropolitan area (MCMA), the largest concentration of compounds detected including methane, were propane and butane. The average concentration of these contaminants was 533 and 250 ppbC, of the 2728 ppbC total, and this corresponds to 19.5% and 9.2%, respectively, Sandoval et al. (2001).

In 1996 there were 3,737,310 residences with LPG stoves, 2,049,531 with LPG heaters and 2,159,379 with clothe washers in the MCMA. This level of demand generates a large LPG consumption, reaching an amount of 67.58 MBD, 31% of the total consumption of fossil fuels, Pemex Gas y Petroquímica Básica (1997). The C_3 and C_4 compounds are emitted to atmosphere in evaporative and exhaust emissions by gasoline and

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Nomenclature

cAI	captive-air irradiation
mBD	miles of barrels per day
NMHC	nonmethane hydrocarbon (ppbC)
NO*x*	nitrogen oxide concentration (ppb)
ppbC	parts per Million of carbon in air
ppb	part per Billion in air
Commercial	commercial liquefied petroleum gas formulation
60% C3/40% C4	synthetic mix 60% propane and 40% n-butane
O_3(max)	maximum ozone level during the day (ppb)

LPG. The emissions by gasoline generate C_4 in larger proportion and the LPG emissions produce C_3 in larger concentrations, Jaimes and Sandoval (2002).

Carter (1994), has determined the incremental reactivity index toward ozone formation for C_3, C_4 and volatile organic compounds (VOCs). Combining the emitted concentration with its applied index value yields the ozone formation potential under a ratio of $NMHC/NO_x = 5$. In Mexico little studies had been carried out to determine this kind of index and no studies have been performed to evaluate the C_3 and C_4 effect on ozone formation under a ratio $NMHC/NO_x = 20$. Because of that, we decided to use smog chambers, which have been successfully utilized to investigate the photochemistry of air pollution by Kelly (1980, 1987) and Gunst and Kelly (1993).

The objective of this study is to determine the LPG influence on ozone formation of MCMA, under natural solar irradiance by additions and dilutions of this kind of energy, and total NMHC of the ambient air. In the dilution experiments with LPG associated compounds, and total nonmethane hydrocarbons, we can determine with certainty, which mixture has the largest impact on ozone formation.

2. Methodology

Captive-air irradiation (CAI) experiments were carried out from November to December of 2000 at the Mexican Petroleum Institute (MPI), located near urban industrial areas with pollutant sources, and it is situated in the trajectory of dominant wind patterns (Norwest of MCMA). The IMP counts on facilities to monitor levels of ozone, nitrogen oxides, oxides of sulfur and carbon monoxide. Transparent bags type A Teflon FEP with 0.051 mm of thickness were used and sealed by means of pressure and heat, each with a 500 liters capacity. Each bag had two Teflon connections of 0.635 cm, one of them is used to fill and evacuate it, and the other one to get samples for analyzers. The Teflon lines were connected to a Pyrex bulb of a 2–liter capacity.

The NO_x and O_3 Thermo Environmental model 42 and 49 analyzers were calibrated several times throughout the study, using a dynamic calibrator coupled to a generator of clean air, and standard gas grade EPA protocol. This system allowed gas dilutions making with +1% precision. The ozone Thermo Environmental model 49-PS photometry calibrator was employed to gauge the ozone analyzer. The Eppley TUVR ultraviolet radiometer, before being used was calibrated in the Center of Sciences of the Atmosphere at the Universidad Nacional Autonoma de México (UNAM). And it was installed closed to the experimental system set up. Type "J" thermocouples were calibrated in the Metrology Laboratory of MPI; it counts on the T-14 accreditation of the National System of Calibration. An aspirated thermocouple was mounted in the control-bag through a 0.635 cm port to measure temperature.

Fig. 1 shows a schematic representation of the experimental system. Each chamber counts on three bulkhead fittings for filling, sampling, and evacuating. The fittings were in the middle of the bottom bags and were linked by Teflon lines to a Pyrex bulb. In order to fill the bags, the 2-l Pyrex bulb was connected to two air compressors in parallel. The filling rate for each bag was nearly identical to this system. To remove some air from a chamber in dilution experiments four vacuum pumps were used in parallel connected to the 2-l Pyrex bulb.

Finally, the addition of a cylinder of ultra-pure air to a 2-l Pyrex bulb after the pumps were disconnected for experiments that required clean air addition. Eight bags were used to carry out the experiments, except that bag nine was used as control-bag. The monitoring instruments were connected to set-up automatic valves lodged in a device to open a specific valve, linked by Teflon line to each chamber to be analyzed. The electric signals from analyzers, temperature and radiometer sensor were sent to the data logger to capture data and display it in real time.

3. Design of experiments

The experimental design consisted on four levels; two reductions below the ambient level (−50% of the total NMHC and −50% of compounds associated to LPG in air), the ambient level, and one level of increase above the ambient level (+60% of LPG formulations). The experimental program was designed with the main objective of finding the effects of different formulations

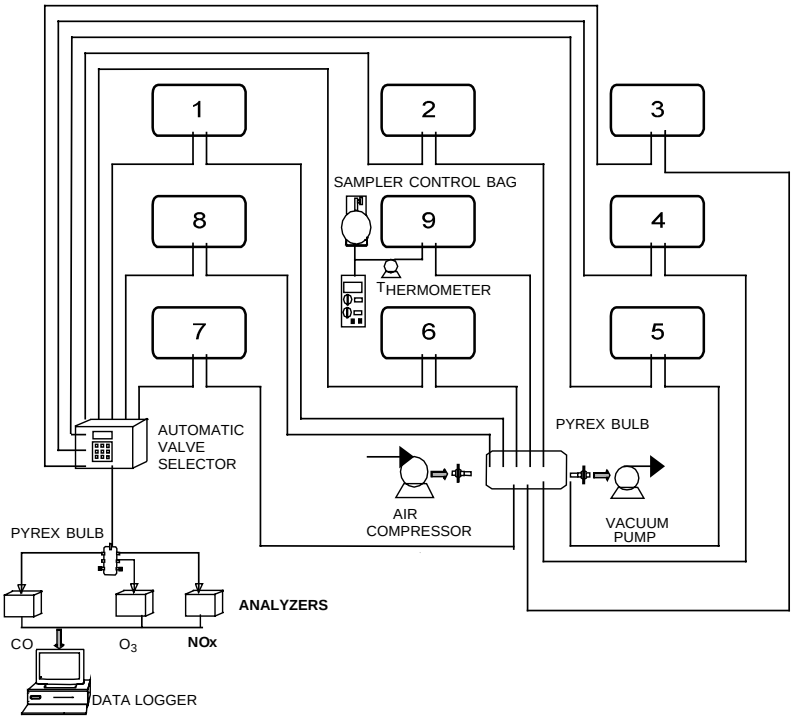

Fig. 1. Experimental system of outdoor smog chambers.

of LPG on O_3 (max) level. A real LPG formulation and one synthesized LPG (Propane–Butane) mixture without olefins 60% C_3/40% C_4 were used.

During the experiments, equal concentrations were added in two different bags to produce replicates (commercial and 60/40). Two bags were used to simulate dilution (subtracting 50% of total NMHC) by removing 250 liters of air from each chamber and refilled with 250 liters of clean air. One of them was injected with a synthetic air mix concentration free of compounds associated with LPG to restore the initial NMHC concentration, and see the effects of decreasing the compounds associated with LPG. Both chambers were injected with NO_x concentration to restore the NO_x initial concentration. In order to compare ozone formation of the six perturbed chambers, two bags were filled with ambient air only (unperturbed chambers).

The eight combination to be tested had the purpose of addressing: (1) if LPG is increased in the air of MCMA, then the ozone levels would be raised; (2) If the compounds associated to LPG were removed from the air of MCMA, the air quality would improve. In order to calculate the ppbC adding concentration of each formulation, the average VOCs/NO_x = 22/1 and the total NO_x morning concentrations measured each day from Mexico City was taken into account, afterwards the bags were filled with ambient air. The formulations and the chambers chosen for each experimental day (ten days each one) were determined randomly.

Each day a sample was taken from the control bag for analyzing VOCs to get to know their concentration. The concentration of the six perturbed bags was calculated with this data.

4. Chamber operation

The experiments began each morning, just before sunrise. First, the chambers were evacuated and filled with 50 liters of ultra-pure air to flush the bags. Then, all bags were filled with ambient air and each bag was prepared according to the experimental design. The chambers were then exposed to natural solar irradiance throughout the day, until sunset. Fig. 2 shows a typical evolution of ozone formation in chamber experiments during the day, filled with ambient air. After sunset, all chambers were evacuated and filled with ultra-pure air, to eliminate contaminants of each bag throughout night to avoid masking the results of the next day.

Fig. 3 shows the temporal O_3 profiles for eight bags of November the 22nd. It can seen that replicates of commercial LPG, 60/40 LPG and unperturbed chambers have very closed values, showing that the experiments give similar results.

For determining the synthetic air mix concentration free of compounds associated with LPG, to be used in experiments, samples of ambient air were collected for five consecutive days in five different sites in MCMA

simultaneously from 6:00 to 9:00 a.m. Sites were: Xalostoc, Merced, Pedregal, Ixtapalapa and Tlalnepantla. Information about five points was obtained, which included four cardinal points and center. Synthetic mixture composition was prepared and contained 11,135 ppb C of VOCs in nitrogen balance, Table 1 shows the concentration.

Due to the fact that the experiments were carried out simulating actual MCMA conditions, and NMHC/NO_x ratio being very important variable; in sites where samples of VOC were taken, NO_x concentration data was also collected. An average ratio of 22/1 was determined. Table 1 also shows LPG commercial, LPG 60/40 and nonmethane Hydrocarbons synthetic mixture concentrations used in experimental runs.

We prepared NO_x mixture for maintaining the same ratio in chambers with dilutions during the experiments as well. This mixture corresponds to 10,000 ppm of NO/NO_x, nitrogen balance and EPA protocol.

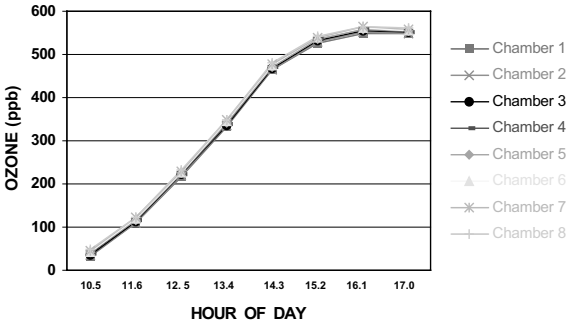

Fig. 2. Temporal O_3 profiles for eight bags replicate experiments.

5. Results and discussion

Table 2 shows initial conditions of experiments, which include the experimentation day in sequence; $NMHC_o$, NO_{xo}, NMHC/NO_x ratio and O_3 (max) formed in two unperturbed chambers, average and maximum temperature.

Fig. 4 shows O_3 (max) effect due to a 60% addition of LPG formulations in ambient air on test days. Two control (unperturbed) bags were included in all experimental days, in order to compare their O_3 (max) with the other bags. This was feasible because perturbed and unperturbed bags were prepared with the same morning air, and exposed to the same natural solar irradiance. This method was used to get the percentage variations of O_3 (max) reported in experiments with ambient air. Commercial formulation showed the larger percent of increasing on O_3 (max), the value was 27%, and 60/40 formulation incremented the O_3 (max) in 21%; these differences in value average are accounted for by the olefins present in commercial formulations and it is well known that these compounds promote ozone formation.

Fig. 5 shows the results of 50% LPG associated compounds dilutions and 50% total NMHC contained in morning ambient air. The 50% associated compounds LPG did not have significant effect, due to the fact that the maximum average ozone had little reduction and was measured at 14%. When the compounds were diminished a 50% total NMHC contained in ambient air, the improvement in air quality was meaningful, diminishing in a 55% the maximum average ozone formed.

Table 3 shows results of experimental runs of twenty days which include nonmethane hydrocarbons (ppbC), nitrogen oxides (ppb) concentration initials and ozone maximum (ppb) in each bag; in this table asterisk (*)

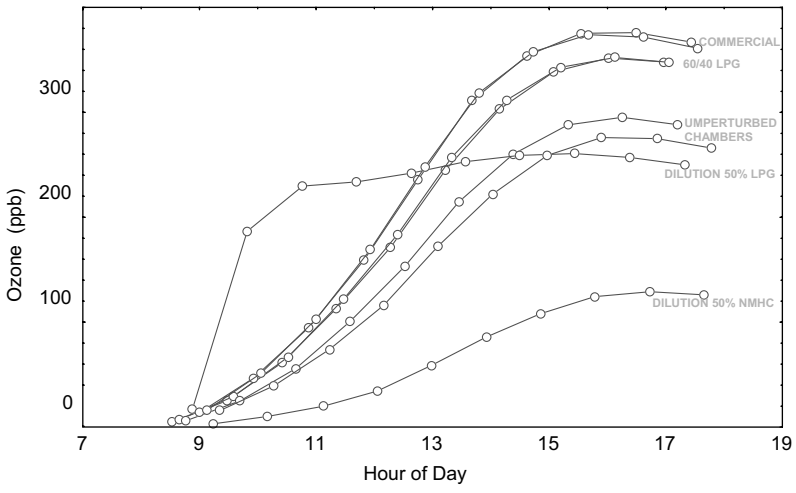

Fig. 3. Temporal O_3 profiles for eight bags of November the 22nd.

Table 1
LPG commercial, LPG and NMHC synthetic mixtures utilized in experimental runs

LPG commercial		LPG 60/40		NMHC	
Compounds	Concentration (ppm)	Compounds	Concentration (ppm)	Compounds	Concentration (ppm)
Propane	6100	Propane	6000	Toluene	722
*n*Butane	2321	*n*Butane	4000	Acetylene	502
*I*butane	1143	—	—	*i*Pentane	465
Ethane	200	—	—	*m*/*p*-Xylene	344
Propylene	124	—	—	—	—
Butene 1	97	—	—	—	—
*i*Pentane	15	—	—	—	—

Table 2
Initial conditions of the experiment and O_3 (max)

Day	NMHCo (ppbC)	NO_{xo} (ppb)	NMHC/NO_x RATIO	O_3 (max) in unperturbed chambers (ppb)	O_3 (max) in unperturbed chambers (ppb)	Temperature average (°C)	Temperature maximum (°C)
22/11/00	1548.3	95	16.30	295.3	273	22.3	26.0
23/11/00	7605.0	330	23.05	643.3	658.7	24.6	28.7
24/11/00	7514.7	257	29.24	681	638.3	23.6	27.3
25/11/00	3791.2	236	16.06	724.3	730.2	24.5	28.3
26/11/00	3036.1	162	18.74	504	519	24.1	27.8
27/11/00	3561.3	280	12.72	865	865	22.7	28.2
28/11/00	2418.6	140	17.28	388	299.3	20.2	24.5
29/11/00	5926.2	220	26.93	691.7	685.7	21.2	25.1
30/11/00	2039.7	128	15.93	308	332	19.5	21.8
01/12/00	3411.0	253	13.48	436.7	480.3	21.2	24.4
02/12/00	2621.5	133	19.71	455.7	445.3	19.3	23.5
03/12/00	2289.1	140	16.35	443.7	442.7	19.6	22.7
04/12/00	1143.6	83	13.78	267.7	258	18.8	20.9
05/12/00	1165.7	65	17.93	250	262	16.6	18.6
06/12/00	2527.4	170	14.87	297.3	239.3	18.9	21.2
07/12/00	2767.3	291	9.51	414.7	396	19.4	23.1
09/12/00	4727.0	362	13.06	500.3	496	22.9	26.0
10/12/00	4842.6	289	16.76	710	686	24.3	28.6
11/12/00	5508.8	430	12.81	598.7	570.7	24.4	27.2
12/12/00	5330.4	225	23.69	694	697	22.8	27.2

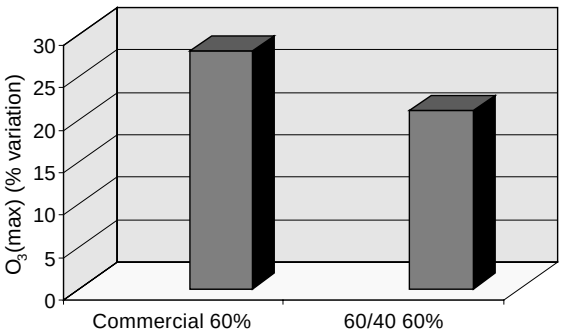

Fig. 4. Effect of addition LPG formulation on O_3 (max) in ambient air.

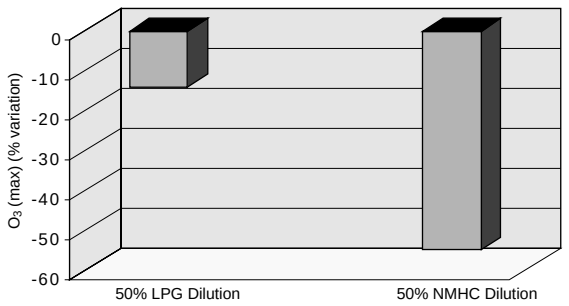

Fig. 5. Effect of 50% compounds associated to LPG and 50% total nonmethane Hydrocarbon dilutions on O_3 (max) in ambient air.

Table 3
Results of runs, nonmethane hydrocarbon (NMHCo), nitrogen oxides (NO_{xo}) concentration initials and ozone maximum (O_3 Max) in each bag

	Run	NMHCo (ppbC)	NO_{xo} (ppb)	O_3 Max (ppb)		Run	NMHCo (ppbC)	NO_{xo} (ppb)	O3 Max (ppb)		Run	NMHCo (ppbC)	NO_{xo} (ppb)	O_3 Max (ppb)
11/22/00					11/23/00					11/24/00				
BAG 1	60/40	2476	93.1	352	BAG 1	B	7605	329	643	BAG 1	D*	3757	229	313
2	60/40	2476	92.3	352.3	2	C	12168	327	920	2	B	7514	253	681
3	B	1548	91.6	295.3	3	B	7605	328	658	3	60/40	12022	251	805
4	D	1548	77.2	260.7	4	60/40	12168	325	866	4	C	12022	249	862
5	C	2476	90.5	376	5	D	7605	315	544	5	60/40	12022	249	805
6	C	2476	90	344	6	60/40	12168	322	814	6	C	12022	246	823
7	D*	774	89.8	129	7	D*	3802	317	178	7	B	7514	231	638
8	B	1548	88.5	276	8	C	12168	320	908	8	D	7514	234	*
	$T_{average}=22.3°C$		$T_{max}=26.0°C$			$T_{average}=24.6°C$		$T_{max}=28.7°C$			$T_{average}=23.6°C$		$T_{max}=27.3°C$	
11/25/00					11/26/00					11/27/00				
BAG 1	C	6065	232	875	BAG 1	C	4857	159	*	BAG 1	C	5697	275	*
2	60/40	6065	230	861	2	B	3036	159	504	2	60/40	5697	275	*
3	B	3791	231	724	3	D	3036	157	409	3	D	3561	266	*
4	B	3791	231	730	4	60/40	4857	158	*	4	C	5697	272	*
5	D*	1895	227	405	5	D*	1518	158	242	5	B	3561	273	737
6	C	6065	229	836	6	C	4857	157	*	6	60/40	5697	269	*
7	60/40	6065	228	816	7	60/40	4857	156	*	7	B	3561	270	865
8	D	3791	211	*	8	B	3036	153	519	8	D*	1780	272	283
	$T_{average}=24.5°C$		$T_{max}=28.3°C$			$T_{average}=24.1°C$		$T_{max}=27.8°C$			$T_{average}=22.7°C$		$T_{max}=28.2°C$	
11/28/00					11/29/00					11/30/00				
BAG 1	60/40	3868	140	388	BAG 1	D*	2963	217	392	BAG 1	B	2039	128	308
2	60/40	3868	139	378	2	B	5926	219	692	2	C	3262	127	441
3	B	2418	138	388	3	B	5926	219	686	3	B	2039	127	332
4	D*	1209	137	171	4	C	9481	218	840	4	60/40	3262	127	416
5	C	3868	137	353	5	C	9481	218	783	5	D*	1019	109	188
6	D	2418	131	338	6	60/40	9481	215	773	6	60/40	3262	125	392
7	B	2418	138	299	7	D	5926	212	491	7	C	3262	126	455
8	C	3868	136	401	8	60/40	9451	213	824	8	D	2039	119	*
	$T_{average}=20.2°C$		$T_{max}=24.5°C$			$T_{average}=21.2°C$		$T_{max}=25.1°C$			$T_{average}=19.5°C$		$T_{max}=21.8°C$	
12/01/00					12/02/00					12/03/00				
BAG 1	C	5457	253	771	BAG 1	D*	1310	131	410	BAG 1	B	2289	140	444
2	60/40	5457	253	674	2	C	4193	131	*	2	60/40	3662	139	499
3	C	5457	251	773	3	B	2621	131	456	3	C	3662	138	505
4	60/40	5457	250	702	4	60/40	4193	129	*	4	D*	1144	122	347
5	D*	1705	250	190	5	B	2621	129	445	5	C	3662	137	495
6	B	3411	250	437	6	60/40	4193	128	*	6	60/40	3662	135	459

7	D	3411	248	*	7	C	4193	128	*	7	B	2289	138	442
8	B	3411	249	480	8	D	2621	122	393	8	D	2289	135	*
	$T_{average}=21.2°C$		$T_{max}=24.4°C$			$T_{average}=19.3°C$		$T_{max}=23.5°C$			$T_{average}=19.6°C$		$T_{max}=22.7°C$	
12/04/00					12/05/00					12/06/00				
BAG 1	D*	571	83	*	BAG 1	B	1165	64	250	BAG 1	B	2527	171	297
2	D	1143	85	*	2	B	1165	64	262	2	D	2527	188	*
3	C	1828	83	298	3	D	1165	54	236	3	60/40	4043	169	385
4	B	1143	83	268	4	C	1864	63	*	4	60/40	4043	166	377
5	B	1143	83	258	5	60/40	1864	62	271	5	C	4043	167	378
6	60/40	1828	82	323	6	D*	582	66	124	6	B	2527	167	239
7	60/40	1828	82	271	7	C	1864	61	*	7	C	4043	166	350
8	C	1828	82	282	8	60/40	1864	61	266	8	D*	1263	169	135
	$T_{average}=18.8°C$		$T_{max}=20.9°C$			$T_{average}=16.6°C$		$T_{max}=18.6°C$			$T_{average}=18.9°C$		$T_{max}=21.2C$	
12/07/00					12/09/00					12/10/00				
BAG 1	C	4427	286	429	BAG 1	C	7563	359	766	BAG 1	D*	7747	294	533
2	B	2767	287	415	2	D	4727	359	749	2	60/40	2421	286	818
3	60/40	4427	285	478	3	60/40	7563	357	686	3	D*	4842	306	795
4	C	4427	279	498	4	D*	1373	366	168	4	B	4842	288	710
5	D*	1383	284	136	5	B	4727	356	498	5	B	4842	286	686
6	D	2767	286	324	6	60/40	7563	352	571	6	C	7747	282	769
7	60/40	4427	282	434	7	B	4727	353	496	7	60/40	7747	283	734
8	B	2767	281	396	8	C	7563	344	624	8	C	7747	281	781
	$T_{average}=19.4°C$		$T_{max}=23.1°C$			$T_{average}=22.9°C$		$T_{max}=26.0°C$			$T_{average}=24.3°C$		$T_{max}=28.6°C$	
12/11/00					12/12/00									
BAG 1	C	8812	418	741	BAG 1	B	5330	225	694					
2	60/40	8812	424	986	2	D*	2665	251	*					
3	D	5508	431	*	3	60/40	8528	222	730					
4	B	5508	424	599	4	60/40	8528	221	757					
5	C	8812	423	674	5	D	5330	250	*					
6	60/40	8812	420	657	6	C	8528	220	*					
7	D*	2754	418	172	7	B	5330	220	697					
8	B	5508	422	571	8	C	8528	218	*					
	$T_{average}=24.4°C$		$T_{max}=27.2°C$			$T_{average}=22.8°C$		$T_{max}=24.9°C$						
						* = Data lost								

Notes:

1. C. Addition of commercial formulation, 60/40. Addition of 60 C_3/40 C_4 formulation, B. Chamber with ambient air unperturbed, D. 50% dilution of compounds associated to LPG, D*. 50% Dilution of total NMHC.
2. Ultraviolet radiation data were lost.

Table 4
Percent of change by 60% addition of LPG formulations and 50% dilution of compounds associated to LPG and total nonmethane hydrocarbons, regarding of unperturbed bags

DAY	Commercial LPG (% of increment on O_3 max)	60/40 LPG (% of increment on O_3 max)	50% dilution of compounds associated to LPG (diminished on O_3 max)	50% Dilution of total NMHC. (diminished on O_3 max)
22/11/00	49.95	25.45	−8.73	−60.08
23/11/00	40.35	34.71	−16.43	−87.0
24/11/00	27.69	30.64	*	−73.24
25/11/00	17.67	24.1	*	−69.82
26/11/00	*	*	−20.04	−65.96
27/11/00	*	*	*	*
28/11/00	9.62	11.61	−1.73	−51.21
29/11/00	17.81	22.35	−28.66	−60.45
30/11/00	40.0	25.21	*	−39.85
01/12/00	68.37	47.1	*	−55.03
02/12/00	*	*	−12.76	*
03/12/00	12.78	12.48	*	−33.57
04/12/00	10.33	11.95	*	*
05/12/00	*	5.29	−7.69	−55.86
06/12/00	35.61	31.2		−35.31
07/12/00	14.35	15.59	−20.07	−83.04
09/12/00	39.54	17.4	*	−44.08
10/12/00	11.03	9.81	*	−20.76
11/12/00	20.97	28.58	*	−49.71
12/12/00	*	6.69	*	*
Average	27.7	21.18	14.5	55.31

* = Lost data.

means a lost data. Some data was lost due to bag leaks and problems with the data acquisition system. After grouping LPG commercial, LPG 60/40, 50% LPG and tNMHC dilution data, the percentage of changes was calculated average of unperturbed bag, shown in Table 4.

6. Conclusions

The addition of 60% LPG commercial formulation was the larger contribution to O_3 (max) formation, compared with LPG 60/40 (propane/butane) formulation added in the same proportion. Ozone formation increased by commercial formulation addition was 28% average; While, 60/40 (propane/butane) presented a 21% average increase.

Regarding the influence of C_3 and C_4 compounds associated to LPG on ozone formation; if it is eliminated a 50% in ambient air of MCMA, O_3 (max) formed diminished 14% only. On the other hand, when a 50% total NMHC in ambient air is eliminated, O_3 (max) formed diminishes 55%.

C_3 and C_4 compounds associated to LPG contribution on ozone formation at the conditions atmosphere, are not meaningful and therefore, they do not cause high ozone pollution.

Regarding Blake and Rowland study, our conclusion complies with the fact that larger C_3 and C_4 concentrations are present in MCMA ambient air; On the other hand, we disagree with the fact that influence C_3 and C_4 means a contribution of up to a 50% in ozone formation, under MCMA conditions.

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