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Impact of the Leakage of Liquefied Petroleum Gas (LPG) on Santiago Air Quality

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Abstract. The leakage of unburned liquefied petroleum gas (LPG) is a major source of urban nonmethane hydrocarbons (NMHCs) in the air of Santiago, Chile. Roughly 5% of the LPG that is sold in Santiago leaks in its unburned form to the atmosphere. Because of the leakage, propane is the most abundant NMHC in Santiago's air, even under heavy traffic conditions. NMHCs are an important precursor to the formation of ground-level ozone, and the LPG leakage may contribute as much as 15% to the excess ozone levels in Santiago. Improvement to the local air quality may be obtained by lowering the rates of LPG leakage, and by minimizing the use of alkene-rich LPG formulations.

Introduction

Santiago is home to 40% of Chile's inhabitants, 800,000 vehicles and 70% of the nation's industries [INE, 1995; Romero *et al.*, 1999]. With a population of 5 million, Santiago is one of Latin America's most polluted cities. The city of Santiago lies in an inland closed basin and is under permanent subsidence and thermal inversion layers [Eskeland, 1997; Romero *et al.*, 1999]. The combination of geographical, meteorological and cultural factors contributes to the extreme air pollution events in Santiago [Romero *et al.*, 1999].

Ozone (O_3) concentrations in Santiago have been found to exceed the 1-hr air quality standard of 80 parts per billion by volume (ppbv) throughout the year, especially in the summer [Ostro *et al.*, 1999 and references therein; Romero *et al.*, 1999]. To reduce ambient O_3 levels, the Chilean Comision Nacional del Medio Ambiente (CONAMA) has recommended that nitrogen oxide (NO_x, = NO_2 + NO) and volatile organic carbon (VOC) emissions in Santiago be halved from 1997 levels by the year 2011. Vehicles and industry have been cited as the most important air pollution sources, and programs are in place to reduce industrial and vehicular pollutant emissions [Romero *et al.*, 1999; www.conama.cl]. However, whereas ethene, ethyne and i-pentane are signature gases for vehicular exhaust and gasoline emissions, our investigation has found that Santiago's air is rich in propane, i-butane and n-butane. Vehicles and industry do not emit large amounts of the C_3-C_4 alkanes [Blake and Rowland, 1995], and a major component of the nonmethane hydrocarbons (NMHCs) in Santiago's air result from the emissions of liquefied petroleum gas (LPG), which is used extensively throughout Santiago for domestic cooking and heating. In this paper we suggest that the impact of LPG leakage on Santiago's air quality needs to be recognized in order for the NMHC reductions to best succeed.

Experimental

The composition of Santiago's air was determined between June 1-8, 1996. A total of 144 whole air samples were collected in a study area 25 km × 21 km that was divided into a 7 × 6 grid and covered most of metropolitan Santiago. Each sample was collected in an evacuated 2-L stainless steel canister that was equipped with a stainless steel bellows valve. With the collaboration of 30 Pontifica Universidad Católica de Chile (PUC) students and faculty, 45 samples were collected throughout the study grid at approximately 5 a.m. on June 4. Minimum vehicular activity was observed at 5:30 a.m., and the 5 a.m. samples represent concentrations before the morning traffic activities had developed. Another 43 samples were collected at the same locations at 9 a.m., after the morning traffic activities had fully evolved.

A comparison of the 5 and 9 a.m. results represents the effects of emissions, chemistry and meteorological transport. In general, the study area had stable atmospheric conditions and weak winds, with a median wind speed of 1.1 km hr^{-1} [Chen *et al.*, 1999]. The rapid daytime boundary layer growth had not begun by 9 a.m., and it is appropriate to compare the mixing ratios measured at 5 and 9 a.m. because the study volumes were similar. The effect of chemistry on the short-lived components of LPG (C_2-C_4 alkenes) is addressed briefly below.

The samples were returned to our laboratory at the University of California – Irvine (UCI) and were analyzed for carbon monoxide (CO), methane (CH_4), 72 NMHCs, 28 halocarbons and 5 alkyl nitrates by gas chromatography (GC) with flame ionization detection (FID) and electron capture detection (ECD). The measurement precision for the NMHCs was 3% or 3 pptv, whichever was larger. Detailed analytical descriptions are given in Hurst [1990] and Blake *et al.* [1996].

Results and Discussion

Composition of Santiago LPG

The three main types of LPG that are used in Santiago ('Propano', 'Butano' and 'Catalytico') were analyzed for 12 C_2-C_5 hydrocarbons by Abastible at their formulation plant. The analysis shows that propane, n-butane, and i-butane are the dominant NMHCs in the Santiago LPG formulations (Table 1). The data also show that Santiago LPG does not contain ethyne, one of the main components of vehicular exhaust.

The percentages that Propano, Butano and Catalytico each contribute to LPG's share of the local NMHC burden in Santiago were calculated to be 79, 16 and 5%, respectively (see below). An average LPG composition for Santiago was determined using these 'burden shares' and the respective compositions of Propano, Butano and Catalytico (Table 1). For example, the average propane content in Santiago LPG (74%; Table 1) is given by multiplying the percentages of

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Table 1. Percentage composition of the three main types of liquefied petroleum gas (LPG) used in Santiago (Propano, Butano and Catalytico). The average LPG composition for Santiago is shown, and the compositions of Mexico City and Los Angeles LPG (based on 1993 and 1995 data) are included for comparison [Blake and Rowland, 1995]. The Santiago LPG was analyzed for 12 C_2-C_5 NMHCs.

Compound	Santiago Propano	Santiago Butano	Santiago Catalytico	Santiago Avg LPG	Mexico City LPG	L.A. LPG
Ethane	0.0	0.0	0.0	0.0	0.0	0.0
Ethene	0.8	0.1	0.0	0.7	0.0	0.0
Ethyne	0.0	0.0	0.0	0.0	0.0	0.0
Propane	92.3	5.2	2.7	73.8	49.0	95.4
Propene	0.7	0.2	1.5	0.7	0.3	1.8
i-Butane	2.9	34.3	28.1	9.2	15.4	1.4
n-Butane	3.2	57.7	15.6	12.6	28.0	0.2
t-2-Butene	0.0	0.1	14.3	0.7	1.2	0.0
1-Butene	0.0	0.1	13.5	0.7	1.0	0.0
i-Butene	0.0	0.1	14.2	0.7	1.0	0.0
c-2-Butene	0.0	0.0	8.6	0.4	1.0	0.0
n-Pentane	0.1	2.2	1.5	0.5	0.4	0.0

propane in Propano, Butano and Catalytico (92.3, 5.2 and 2.7%, respectively) by their respective burden shares (79, 16 and 5%), and summing the resulting products.

Propane is clearly the dominant component in the averaged Santiago LPG (Table 1). I-butane and n-butane together contribute 22% to the averaged LPG, whereas C_4 alkenes are present in large quantities only in the Catalytico formulation. Because Catalytico represents just 5% of the LPG burden share, the four C_4 alkenes listed in Table 1 each comprise less than 1% of the averaged LPG composition. Even though the alkenes are present in such minor quantities, they are nevertheless very effective in their contribution to local O_3 production (see below).

Rate of LPG Leakage in Santiago

Santiago's air contained extremely high concentrations of many hydrocarbons and some halocarbons when compared to background samples collected at the coast near San Antonio, Chile. For example, the concentration of propane was more than 1500 times higher in Santiago than near San Antonio. On June 4, the median propane mixing ratios at 5 and 9 a.m. (131 and 136 ppbv, respectively) accounted for 42 and 28% of the total measured NMHC burden in Santiago. Indeed, propane is the most abundant NMHC in Santiago's air, even under the heavy traffic conditions at 9 a.m. (Figure 1). We recall that propane is the dominant component in Santiago LPG (Table 1) and the high propane levels suggest significant leakage of LPG in the unburned form.

The daily rate of LPG leakage in Santiago was calculated as follows. First, an average study volume of roughly 320 km^3 was calculated using the study area of 25 km × 21 km multiplied by an estimated diurnally averaged boundary layer (BL) height of 600 m. At 9 a.m. on June 4 the BL height was visually estimated to be 200 ± 100 m [Chen *et al.*, 1999], suggesting a slow evolution of the daytime convective BL. The diurnally averaged value of 600 m is based on a BL height of 200 m for 14 hours, and 1200 m for 10 hours (see also Kaimal and Finnigan, 1994). Second, a one-day ventilation of the study volume was estimated based on 43 surface wind observations on June 4 that gave a median wind speed of 1.1 km hr^{-1}. This estimate agrees well with wind speed observations from CONAMA monitoring stations, which in turn support a one-day ventilation of the study volume. Third, the median propane, n-butane and i-butane mixing ratios for all Santiago city samples collected between June 1-8 were 137.5, 27.0 and 16.1 ppbv, respectively (Figure 1). Recalling that the background concentrations of the LPG components were negligibly small compared to the Santiago mixing ratios, and assuming that each measured median mixing ratio is representative of a typical diurnal value, these mixing ratios correspond to burdens of 80, 20 and 12 tons of propane, n-butane and i-butane, respectively, leaking daily into the study volume. The remaining components of LPG add another 6 tons to the daily NMHC burden, for a total daily burden of roughly 120 tons resulting from LPG leakage. Finally, the amount of LPG sold daily in Santiago is about 2500 metric tons [private conversation with industry representatives, 1996]. From the above we calculate a daily LPG leakage rate of roughly 5%. We estimate that the LPG leakage rate could range from 3-6%, based on uncertainties in the diurnally averaged BL height and the daily ventilation rate. By comparison, we determined that uncertainty in the NMHC burdens, including the effect of chemistry on the short-lived components of LPG (C_2-C_4 alkenes), introduces negligible uncertainty to the calculated LPG leakage rate.

Contribution of LPG to the NMHC Composition of Santiago's Air

We recall that the principal components of LPG do not have significant industrial or vehicular sources. That is, LPG is the primary source of the C_3-C_4 alkanes in the air of Santiago. By contrast, the other known components of LPG (C_2-C_4 alkenes and n-pentane) are also strongly influenced by sources such as vehicular usage. To determine contribution of LPG leakage to ground-level O_3 production in Santiago, we need to first calculate the percentage that LPG contributes to the ambient mixing ratios of n-pentane and each C_2-C_4 alkene (Pi_{LPG}). To this end, air samples were collected on June 6, 1996 both in metropolitan Santiago and inside the 2 km long Zapata tunnel. The Zapata samples were used to estimate the emissions from vehicular traffic traveling inside the tunnel.

The percentages that Propano, Butano and Catalytico contribute to the measured burden of all 12 NMHCs listed in Table 1 (P_{Pro}, P_{But} and P_{Cat}, respectively) were used to determine Pi_{LPG} for the C_2-C_4 alkenes and n-pentane. P_{Pro}, P_{But}

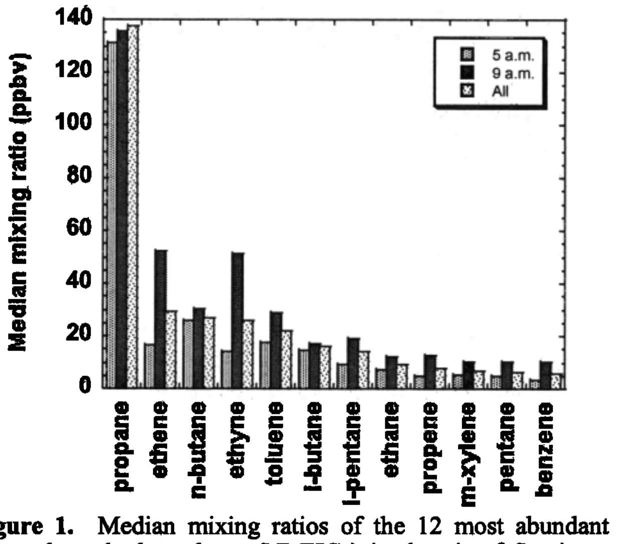

Figure 1. Median mixing ratios of the 12 most abundant nonmethane hydrocarbons (NMHCs) in the air of Santiago. The 5 and 9 a.m. samples were collected on June 4, 1996; 'All' denotes median mixing ratios for all Santiago city samples collected from June 1-8. Mixing ratios of compounds with LPG as their major source (propane, i-butane, n-butane) changed little between 5 and 9 a.m., whereas compounds with a strong vehicular source (e.g. ethene, ethyne) showed a large increase as the morning traffic activities fully developed.

Table 2. Production of O_3 in Santiago by CO, CH_4 and 24 NMHCs (in descending order), June 1-8, 1996. The 10 known components of LPG are indicated in bold (alkanes) and bold italics (alkenes). Mole fraction (*MF*) was determined using the median mixing ratios of CO, CH_4 and 43 NMHCs; *ROP* is the relative O_3 productivity; Pi_{LPG} is the percentage that each LPG component *i* contributes to its concentration in Santiago's air; O_3 production by LPG is the O_3 production times Pi_{LPG} divided by 100; *MIR* is maximum incremental reactivity; *POCP* is the photochemical ozone creation potential.

Compound	*MF* (×100)	*ROP*	O_3 production (%)	Pi_{LPG} (%)	O_3 production by LPG (%): MIR scale	O_3 production by LPG (%): POCP scale
m-xylene	0.12	869	10.8			
Ethene	***0.52***	***207***	***10.7***	***4***	***0.4***	***0.4***
Toluene	0.38	248	9.5			
CO	55.14	2	8.4			
1,2,4-TMB	0.05	1056	5.7			
Propene	***0.14***	***395***	***5.5***	***16***	***0.9***	***0.8***
propane	**2.41**	**21**	**5.1**	**100**	**5.1**	**13.9**
o-xylene	0.07	689	4.6			
p-xylene	0.06	700	4.0			
1,3,5-TMB	0.03	1212	3.1			
n-butane	**0.47**	**59**	**2.8**	**100**	**2.8**	**7.2**
i-pentane	0.25	99	2.5			
t-2-butene	***0.04***	***560***	***2.5***	***53***	***1.3***	***1.1***
c-2-butene	***0.04***	***560***	***2.2***	***36***	***0.8***	***0.7***
1-butene	***0.04***	***481***	***2.0***	***52***	***1.1***	***1.0***
i-butane	**0.28**	**68**	**1.9**	**100**	**1.9**	**3.6**
2Me2Butene	0.04	448	1.9			
1,3-butadiene	0.03	589	1.6			
1,2,3-TMB	0.01	1068	1.5			
ethylbenzene	0.05	286	1.5			
i-butene	***0.05***	***286***	***1.4***	***48***	***0.7***	***0.6***
2-MePentane	0.08	129	1.1			
Mechexane	0.05	180	1.0			
CH_4	38.34	0.2	0.9			
1-pentene	0.02	434	0.9			
n-pentane	**0.12**	**75**	**0.9**	15	0.1	0.4

	MIR scale	POCP scale
LPG **alkane** contribution to O_3 production	10.0%	25.1%
LPG ***alkene*** contribution to O_3 production	5.2%	4.6%
Total LPG contribution to O_3 production	**15.2%**	**29.6%**

and P_{Cat} were determined by minimizing the sum of the 12 residuals of:

$$MFi_{San} - (MFi_{Zap} \times P_{Zap}) - [(MFi_{Pro} \times P_{Pro}) + (MFi_{But} \times P_{But}) + (MFi_{Cat} \times P_{Cat})] \quad (1)$$

where Eq. (1) was applied to each of the 12 compounds listed in Table 1 (*MF* is the measured mole fraction for a given NMHC *i*, for Santiago (MFi_{San}), Zapata tunnel (MFi_{Zap}), Propano (MFi_{Pro}), Butano (MFi_{But}) and Catalytico (MFi_{Cat}); $P_{Zap} = 100\% - [P_{Pro} + P_{But} + P_{Cat}]$). The minimized sum of the 12 residuals was 3.8%, and P_{Pro}, P_{But} and P_{Cat} were found to be 54.4, 11.1 and 3.4%, respectively, for a total contribution to the burden of these 12 NMHCs of 69% by LPG and 31% from vehicles. The low residual sum supports that LPG and vehicles are the principal sources of these 12 compounds, and that industry contributes minimally to their ambient mixing ratios. We also used a Principal Component Analysis (PCA) to confirm that LPG and vehicles explain most (92%) of the variance among these 12 compounds.

We use trans-2-butene as an example to show how P_{Pro}, P_{But} and P_{Cat} were used to determine Pi_{LPG}. The percentages of trans-2-butene in Propano, Butano and Catalytico are 0.0, 0.1 and 14.3%, respectively (Table 1). From above, P_{But} and P_{Cat} are 11.1 and 3.4%, respectively. Therefore, the leakage of trans-2-butene from Butano and Catalytico contributes (0.001 × 11.1%) + (0.143 × 3.4%) = 0.50% to the median NMHC concentration in Santiago's air, again for the 12 compounds listed in Table 1. By comparison, trans-2-butene (from both LPG and other sources) comprises 0.94% of the NMHC composition of Santiago's air for the same 12 compounds (data not shown). Therefore, the contribution of LPG to the total mixing ratio of trans-2-butene in Santiago's air is 0.50 ÷ 0.94 = 53%. Table 2 shows the contribution of each LPG component to its overall mixing ratio in Santiago's air.

To calculate the percentages that Propano, Butano and Catalytico each contribute to LPG's share of the local NMHC burden, P_{Pro}, P_{But} and P_{Cat} were respectively divided into their sum of 68.9%. Thus for Propano, 54.4 ÷ 68.9 = 79%, and Propano is responsible for 79% of the NMHC burden in Santiago resulting from LPG leakage. Similarly, Butano and Catalytico comprise 16 and 5% of LPG's share, respectively.

Contribution of LPG to Excess O_3 Production in Santiago

The leakage of unburned LPG contributes significantly to the production of ground-level O_3 in Santiago. Ground-level O_3 is created in the reactions of excess concentrations of NMHCs and NO_x in the presence of sunlight (Reactions 2-6, where R is an alkyl group; '·' denotes a radical; $h\nu$ is a photon; and M is a third-body molecule):

$$RH + OH\cdot \rightarrow R\cdot + H_2O \quad (2)$$

$$R\cdot + O_2 \rightarrow RO_2\cdot \quad (3)$$

$$RO_2\cdot + NO \rightarrow RO\cdot + NO_2 \quad (4)$$

$$NO_2 + h\nu \rightarrow NO + O \quad (5)$$

$$O + O_2 + M \rightarrow O_3 + M \quad (6)$$

The subsequent reactions of various RO· species created from Reaction 4 are also important for O_3 production [Blake and Rowland, 1995]. The contribution of each NMHC to local O_3 production can be approximated based on the maximum incremental reactivity (MIR) estimates of Carter [1994]. A compound's MIR multiplied by its molecular weight gives the relative O_3 productivity for that compound (ROP_i; Blake and Rowland, 1995). The per molecule O_3 production for each compound is then given by $ROP_i \times MF_i$, divided by the sum of $ROP_i \times MF_i$ for all the compounds. Each compound's *MF* was calculated as its median mixing ratio (from all Santiago city samples collected between June 1-8) divided by the sum of the median mixing ratios for CO, CH_4 and the NMHCs (Table 2).

The contributions of propane, n-butane and i-butane to O_3 production in Santiago are 5.1, 2.8 and 1.9%, respectively, for a total contribution of 9.9% (Table 2). This contribution is based on LPG as the sole source of the C_3-C_4 alkanes in the local air. By contrast, the contribution of the C_2-C_4 alkenes and n-pentane from LPG leakage to O_3 production is given by the per molecule O_3 production multiplied by the percentage that LPG contributes to the ambient concentration of each molecule (Pi_{LPG}; see above). For example, the contribution of trans-2-butene from LPG to O_3 in Santiago is (2.5% × 0.53) = 1.3% (Table 2). Together, C_2-C_4 alkenes and n-pentane from LPG contribute 5.3% to the local O_3 production, and the 10 known components of LPG are estimated to contribute 15% to the production of O_3 in Santiago.

The MIR estimates are used to calculate O_3 production in the urban airshed in which the NMHC emissions occur [Carter, 1994]. On a regional scale, the contribution of each NMHC to O_3 production can be calculated using the photochemical ozone creation potentials (POCPs) of Derwent *et al.* [1998]. Scaled to ethene, the POCP method yields larger O_3 production for the simple alkanes, and smaller production for the light alkenes than the MIR scale (Table 2). Based on the POCP values, the contribution of Santiago's LPG leakage to regional O_3 production is estimated to be on the order of 30% (Table 2). When applying the MIR and POCP scales, it is important to recognize that neither perfectly fits the Santiago situation, and we use these calculations to assess the general significance of our Santiago observations.

The contribution of LPG to O_3 production in Santiago can be compared to rates in other urban areas (Table 1). Compared to Santiago, the Mexico City LPG formulation was found to be relatively lower in propane (50-60%), and higher in butanes (35-45%) and alkenes (3-5%) [Blake and Rowland, 1995; Gamas *et al.*, 2000]. That is, the Mexico City formulation has been in the direction of the more reactive butanes and alkenes. As a result, the unburned leakage and incomplete combustion of LPG have played a major role in O_3 production in Mexico City [Blake and Rowland, 1995]. Indeed, the ventilation of LPG components from the Valley of Mexico may contribute 0.5% to the northern hemispheric inputs for both propane and n-butane [Elliott *et al.*, 1997]. By contrast, the cumulative O_3 productivity for Los Angeles LPG is roughly half that of Mexico City, because of both a lower alkene content and a switch in the C_3-C_4 composition towards the less reactive propane [Blake and Rowland, 1995]. The identification of LPG as a major precursor to O_3 formation in Mexico City has led to legislation that limits the alkene content in Mexican LPG [Blake and Rowland, 1995; Gamas *et al.*, 2000]. Similarly, the air quality in Santiago would benefit from switching the LPG towards a formulation that contains lower amounts of the more reactive hydrocarbons.

Conclusions

Leakage of LPG is a leading source of NMHCs in Santiago's air, and propane is the dominant NMHC in Santiago's air, even under heavy traffic conditions. The C_3-C_4 alkanes from LPG contribute approximately 10% to the O_3 formation in Santiago. Although alkenes are a minor component of LPG, they furnish another 5% to local O_3 production. Improvement to both local and regional air quality may be obtained by minimizing LPG leakage, for example by periodically checking and replacing O-rings and seals. Additional improvement may be possible by phasing out the alkene-rich Catalytico formulation.

Programs are currently in place in Santiago to reduce industrial and vehicular pollutant emissions. In addition, Santiago is undergoing a conversion from coal and oil to natural gas. We suggest that implementing the above recommendations for reducing LPG leakage would complement the current efforts to improve Santiago's air quality. In a future study we plan to revisit our sampling grid in Santiago in order to investigate the impact of the pollutant reductions, the switch to natural gas, and any changes in LPG composition or handling on the local air quality.

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