Analysis of the Influence of Sources on the Spatial Variation in Atmospheric Methane Concentrations in the City of Tandil, Argentina

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Abstract

There is an overall trend in urban methane (CH_4) emissions due to the presence of several sources; however, differences exist between cities, and therefore further local research should be undertaken. The present study analyzes the spatiotemporal variation in atmospheric CH_4 concentrations during a year at ten sampling sites in the urban core of a medium-sized city. The mean annual atmospheric CH_4 concentrations varied between 2.02 ppm and 5.45 ppm; the maximum concentrations were found in a site close to a wastewater treatment plant (WWTP), presenting a significant increase toward the summer. In the rest of the sites, the maximum concentrations were recorded in the coldest months due to the influence of combustion sources dependent on natural gas (NG). An exploratory regression analysis was performed, in which the variables “homes connected to the gas network” and “distance from compressed NG stations” each explained 66 and 65% of the spatial variation of the atmospheric CH_4 concentrations at the 9 sites (excluding that one nearest the WWTP). The results show the need to prevent NG leaks in all urban areas to reduce the emissions of this potent greenhouse gas, which, at the same time, will provide economic benefits for the sectors involved.

Introduction

The task of reducing greenhouse gases (GHGs) emissions sufficiently to stabilize their atmospheric concentrations and therefore mitigate climate change is a great global challenge (Erickson 2017). Methane (CH_4) becomes relevant because its atmospheric concentrations continue to increase, making it the second most important anthropogenic GHG in terms of climate forcing, after carbon dioxide (CO_2) (Saunois et al. 2020).

CH_4 emission reduction is technically challenging: it demands identifying each source, quantifying its emission flux, and developing effective emission reduction methodologies (Nisbet et al. 2020). CH_4 is emitted by a variety of processes, including natural and anthropogenic ones, and by a variety of sources, fixed and diffuse, non biogenic and biogenic, thermogenic and pyrogenic ones (Saunois et al. 2020). The goal should be to reduce anthropogenic emissions, which we can control (Nisbet et al. 2019).

Cities generate 70% of anthropogenic GHGs emissions, a fraction that is growing with global urbanization (Hopkins et al. 2016). Because of their density, efficiency, and adoption of innovations and new technologies, cities can provide solutions for reducing emissions (Hoornweg et al. 2011). The effectiveness of these remedial actions depends on accurate knowledge of the many sources of GHG in each city (Ars et al. 2020). In many cities, estimating the source contribution when so many emission sources coexist, can be challenging. Besides, according to Nisbet et al. (2020), the location of emissions may be complex because what initially appears to be a point source may actually comprise many small sub-sources.

In general, the urban sources of GHGs can be classified in energy (electricity, heating), industry (processes, product use), transportation (road, non-road, navigation, take-offs, landings, and aircraft flying

over urban areas), and waste (landfills, wastewater treatment) (Marcotullio et al. 2013). Energy and transportation emit primarily fossil CH_4 derived from natural gas (NG) (Balcombe et al. 2017; Cai et al. 2017; Bo Zhang et al. 2014), whereas waste treatment produces biogenic CH_4 from the process of anaerobic decomposition (El-Fadel and Massoud 2001). Leaks in the NG distribution network and low efficiency during gas use (for heating, for cooking, or as vehicle fuel) contribute greatly to anthropogenic CH_4 emissions (Fusé et al. 2019; Hendrick et al. 2016; Hopkins et al. 2016, Hu et al. 2018, Saint-Vincent and Pekney 2019; Wong et al. 2016). Compared with other fuels, compressed NG (CNG) is economically attractive, and as a result, the number of CNG vehicles has notably increased in the past years (Kumar et al. 2015). In the waste sector, CH_4 emissions depend mainly on the waste generated (its kind and volume), waste separation and disposal, the number of homes, and the socioeconomic level of the population (Ranieri et al. 2017; Yan et al. 2019; Zhao et al. 2019).

GHGs emissions are significantly associated with population size, density, growth rates, and per capita income (Marcotullio et al. 2013). At present, cities are undergoing a profound transformation that especially affects the territorial structures of their peripheries (Palomares and Puebla 2007; Linares and Picone 2018). The expansión of urban peripheries gradually results in population decentralization, and separation between daily activities and access to services (Rufí 2003). This urban expansion is accompanied by the emergence of new pollution emission sources (Hien et al. 2020), which causes significant spatial and temporal variations in the atmospheric concentration of contaminant gases (Hien et al. 2020; Norman et al. 2006). The development and application of models that allow quantifying the interrelationships between environmental and socioeconomic factors associated with urban growth will help us to learn about the possible future effects of their evolution, key to preventing associated problems (Linares 2019). Although there are overall trends in urban CH_4 emissions, differences exist between cities (geophysical factors—climate, access to resources—and technical factors, such as power generation, urban design, waste processing), which need to be explored by further local research (Kennedy et al. 2009).

In Argentina, some authors have conducted studies on GHG emissions in specific urban sectors: waste (Sanci and Panarello 2016), NG residential use (Fusé et al. 2019), and transport (Puliafito et al. 2015). In general, the GHG inventories that Argentina submits to the United Nations Framework Convention on Climate Change (UNFCCC) secretariat, following IPCC guidelines, are based on data and emission factors that do not apply to this region (Secretariat of the Environment and Sustainable Development 2015). This leads to considerable uncertainty about the national GHG emission budget.

The aim of this study is to identify possible CH_4 sources in a medium-sized city in order to explain the spatial variability of atmospheric CH_4 concentrations. For it, periodical measurements were performed in several strategically selected urban sites, and then, using spatial analysis tools, the relative contribution of CH_4 from each source was evaluated.

Materials And Methods

The Study Site

Tandil city (37°19' S, 59°08' W; with an area of 52.34 km^2 and a population of 116.916) is located in the southcenter of Buenos Aires province, Argentina. To measure atmospheric CH_4 concentration, 10 sampling sites were selected (Fig. 1a), ensuring a uniform distribution over the urban area that would include different urban densities, residential and commercial uses of soil (Figs. 1b and 1c), and CH_4 sources (Fig. 2).

For the meteorological characterization of the study period, data regarding air temperature, relative humidity, atmospheric pressure, visibility, and wind velocity were obtained daily from the National Meteorological Service, Tandil-AERO weather station. In addition, information on NG monthly consumption—CNG, residential, commercial, and public consumption by different sectors of the city was provided by the only NG supplier (CAMUSSI S.A).

Monitoring of atmospheric CH_4 concentrations

During a study period of one year (June 1st, 2017 – May 31st, 2018), integrated air samples were obtained in consecutive 15-day periods. The samples were collected in 0.5 liter stainless steel canisters with an inlet valve (Fusé et al. 2019) and an air flow controller calibrated for the desired sampling period (Gere and Gratton 2010). The canisters were previously cleaned with high purity nitrogen and evacuated to approximately 0.5 mb pressure. At the end of each sampling period, the canisters were replaced and transferred to the laboratory, where samples were analyzed for CH_4 concentration by gas chromatography (CG Agilent 7890A) (Fusé et al. 2019). Regarding measurement error, the chromatograph ranges from 1% to 3% (0.02 to 0.05 ppm for ambient air).

Data analysis and statistics

Basic descriptive statistical analyses were performed to assess temporal and spatial variation in atmospheric CH_4 concentrations measured in each sampling site. An ANOVA Test and a Fisher's LSD test were performed to examine temporal differences between atmospheric CH_4 concentrations measured in each site and analyze the differences between sampling sites (mean values with the same letter are not statistically different). The relative incidence of biogenic and non biogenic sources in the atmospheric CH_4 concentrations at each site was measured using the Pearson correlation analysis with ambient temperature and with NG sectoral consumption. For all the analyses a p-value of 0.05 was considered to assess significance. Infostat Statistical software was used for all statistical analyses.

Exploratory Regression

Spatial analysis tools (ArcGIS 10.5®) were used to identify the independent variables that account for the mean CH_4 values obtained. First, georeferencing of the sampling sites was performed. To assess the influence of the environment on each sampling site, a buffer zone (area of influence) was created around

each one (Hoek et al. 2008). We selected a buffer radius of 300 m, following the recommendations by several authors (Briggs et al. 2000; Henderson et al. 2007).

Based on the last National Census of Population and Housing in the city of Tandil (INDEC 2010), we calculated the average of homes connected to the NG network (GN) within each census tract that included one of the buffers at the sampling sites (Fig. 2a). In addition, the fixed sources that could a priori contribute to the spatial variation in atmospheric CH_4 concentrations in the urban area—CNG stations, a wastewater treatment plant (WWTP) and an artificial lake, were represented in a vector layer, and the Euclidean distance to each sampling site was calculated. In this way, three individual maps were created: distance in meters from the CNG station (GD), from the WWTP (PD), and from the artificial lake (LD) (Figs. 2b, 2c and 2d).

Taking into account that the dependent variable is the mean CH_4 concentrations in each site (seasonal and annual) and that the independent variables are the four layers created, an exploratory regression analysis was performed. This tool examines whether the association between variables is constant in the whole urban area or whether variations take place with the goal of looking for the Ordinary Least Squares (OLS) models that best explain the dependent variable (Habermann et al. 2015; Bertazzon et al. 2015). Such a relationship can be positive or negative in sign (directly or inversely proportional) with a high or low % of significance of the variable (consistency in the relationships). To validate the results (*passing models*), the coefficients considered were the following: the adjusted R^2 (Adj R^2) > 0.50, the level of significance $p < 0.05$, the Variance Inflation Factor (VIF, that indicates independence between the independent variables) < 7.5, the spatial autocorrelation I Global Moran with $p > 0.10$ and the residual normality summary Jarque-Bera with $p > 0.10$ (Buzai and Baxendale 2012; Quinn and Keough 2002; Rosenshein et al. 2011). It should be noted that for all OLS indices a significance level of $p < 0.01$ was used.

Results And Discussion

Meteorological parameters and natural gas consumption

Over the study period, mean monthly temperature was 14.6°C, with a variation range from 8.5 to 21.4°C. Regarding humidity, air pressure and visibility the measured mean values were 71.0 %, 1015.4 hPa and 15.1 Km, respectively. Average total monthly rainfall was 78.1 mm. Precipitation and air temperature presented the highest temporal variability (coefficient of variation equal to 32.2 % and 42.3 %, respectively). Whereas precipitation showed no statistically significant differences between the 4 seasons ($p > 0.05$), temperature did show differences ($p < 0.05$) except in the fall and the spring. Wind intensity remained almost constant (mean monthly equal to 13.5 Km h^{-1}) throughout the period and showed no statistically significant differences between seasons ($p > 0.05$), except for the fall.

Fig. 3 shows the temporal variation in monthly NG consumption in the city of Tandil for the residential, commercial, and CNG sectors during the study period. Of the total NG consumption in the city of Tandil,

65.2 %, 10.6 %, 10.0 % and 14.1% correspond to the residential, commercial, CNG and industrial (not considered in this study) sectors, respectively. In Argentina, the highest percentages of consumption of NG distributed along the gas network correspond to the residential and industrial sectors—43.5 % and 39.7 %, respectively (Secretary of Energy of the Nation 2019).

NG residential and commercial consumption are higher in winter, with statistically significant differences in relation to the other seasons in the case of the residential sector and to spring and summer in the case of the commercial sector ($p < 0.05$). This behavior was reflected in a good correlation between NG consumption and ambient temperature, with an R and p value (value in parenthesis) of -0.78 ($p = 0.003$) and -0.94 ($p < 0.0001$) for the residential and commercial sectors, respectively. In general, heating degree days is currently an important determinant of the amount of energy required to heat urban buildings (Kennedy et al. 2009). In our country, residential and commercial NG consumption reaches its maximum level in the winter months (Secretariat of the Environment and Sustainable Development 2015).

Regarding CNG demand, only statistically significant differences were measured between summer and winter ($p < 0.05$), with a fairly good correlation with ambient temperature ($R = -0.69$, $p = 0.013$). A decrease in the CNG consumption in the summer is probably related to less economic activity as a consequence of the holidays (Gil 2006; Gioli et al. 2012).

Temporal variability of atmospheric methane concentrations

Table 1 shows the average atmospheric CH_4 concentrations for each season and for the complete study period at each sampling site.

Table 1 Comparison of the mean values of the atmospheric CH_4 concentrations between seasons (± standard deviation, n = total number of measurements performed in each site, superscript letters show results for Fisher's LSD Test) and results of Pearson correlation analysis (linear correlation coefficients R and significance level p in parentheses) between the atmospheric CH_4 concentrations and the mean air temperature (T) at each site

Site	Atmospheric CH_4 concentration (ppm)					*n*	Pearson [CH_4] with T
	Spring	Summer	Fall	Winter	***Annual***		*R (p)*
1	2.14 ± 0.05^{a}	2.08 ± 0.04^{a}	2.26 ± 0.06^{ab}	2.45 ± 0.37^{b}	***2.22 ± 0.23***	20	-0.71 (< 0.001)
2	2.16 ± 0.04^{a}	2.12 ± 0.07^{a}	2.36 ± 0.12^{b}	2.38 ± 0.06^{b}	***2.25 ± 0.14***	20	-0.92 (< 0.001)
3	4.24 ± 1.03^{a}	5.78 ± 1.07^{b}	6.18 ± 2.03^{b}	5.58 ± 0.45^{ab}	***5.45 ± 1.41***	20	-0.24 (0.455)
4	2.09 ± 0.07^{a}	2.08 ± 0.04^{a}	2.30 ± 0.17^{b}	2.27 ± 0.04^{b}	***2.19 ± 0.14***	20	-0.82 (0.001)
5	2.16 ± 0.04^{a}	2.09 ± 0.02^{a}	2.30 ± 0.09^{b}	2.30 ± 0.08^{b}	***2.21 ± 0.11***	24	-0.93 (< 0.001)
6	2.19 ± 0.11^{a}	2.12 ± 0.04^{a}	2.31 ± 0.14^{a}	2.57 ± 0.32^{b}	***2.30 ± 0.24***	24	-0.78 (0.003)
7	2.05 ± 0.05^{a}	2.01 ± 0.05^{a}	2.22 ± 0.24^{b}	2.12 ± 0.07^{ab}	***2.10 ± 0.14***	24	-0.65 (0.023)
8	2.32 ± 0.09^{ab}	2.22 ± 0.07^{a}	2.46 ± 0.17^{b}	2.45 ± 0.09^{b}	***2.37 ± 0.14***	23	-0.66 (0.020)
9	2.11 ± 0.02^{a}	2.09 ± 0.05^{a}	2.26 ± 0.10^{b}	2.31 ± 0.16^{b}	***2.19 ± 0.13***	24	-0.82 (0.001)
10	2.04 ± 0.04^{b}	1.97 ± 0.03^{a}	2.03 ± 0.04^{b}	2.05 ± 0.04^{b}	***2.02 ± 0.05***	22	-0.79 (0.003)

Except for S3 (the one nearest the WWTP and located in a high-density zone), at all sites the greatest CH_4 concentrations were registered in the winter and/or the fall, with significant statistical differences with respect to the other seasons. A good inverse statistically significant correlation was obtained between the mean monthly atmospheric CH_4 concentration at each site and monthly mean temperature (Table 1). Given that the maximum CH_4 concentrations were recorded in the coldest months, we infer that the predominant sources are non biogenic ones, and not sources associated with biological processes, whose emissions occur in the warm seasons (Wong et al. 2016).

Fusé et al. (2019) suggested that the maximum concentrations of atmospheric CH_4 recorded in the fall and winter in Tandil can be explained by the higher consumption of NG for heating and by gas leaks in the heating systems. In general, good statistically significant correlations were obtained between

atmospheric CH_4 and residential or commercial consumption of NG (Table 2). NG exhaust from diverse residential implements (for heating, water heating, and cooking) contains some unburned CH_4 due to inevitable incomplete combustion (Lebel et al. 2020; Merrin and Francisco 2019). Helfter et al. (2016) suggest that in winter, increases in CH_4 concentrations above the background level could be attributed to CH_4 losses from over-pressurized pipelines as a response to an increase in gas demand.

Table 2 Linear correlation coefficients R and significance level p (in parentheses) between the values of atmospheric CH_4 concentration for each site and the NG volumes for each sector

	Sector		
Site	Residential	Commercial	CNG
1	0.71 (0.01)	0.70 (0.01)	0.43 (0.16)
2	0.58 (0.05)	0.93 (< 0.01)	0.57 (0.05)
3	-0.13 (0.70)	0.33 (0.29)	0.12 (0.70)
4	0.48 (0.11)	0.77 (< 0.01)	0.29 (0.35)
5	0.61 (0.04)	0.92 (0.03)	0.57 (0.05)
6	0.75 (0.01)	0.79 (0.01)	0.58 (0.05)
7	0.30 (0.33)	0.58 (0.05)	0.08 (0.80)
8	0.45 (0.14)	0.63 (0.03)	0.51 (0.09)
9	0.67(0.02)	0.84 (< 0.01)	0.33 (0.29)
10	0.68 (0.02)	0.62 (0.03)	0.44 (0.15)

A better correlation between atmospheric CH_4 and commercial consumption of NG was obtained for S2, S4, S5 and S9, probably because these sites are located in commercial areas or close to them (La Macchia 2016). S9 is situated at one of the main entrances to the city, where various stores are located (Migueltorena and Linares 2019).

The best correlation between atmospheric CH_4 and CNG demand was observed in S6. This site is located near National Route 226 and two CNG stations (Fig. 2b). A good correlation was also established for other sites close to CNG stations, S2, S5, and S8, although the last one with a value of $p < 0.1$. In the CNG stations, CH_4 emissions may result from the process of converting pipeline gas to vehicle fuel and during the fueling process itself. Recent surveys provide evidence for fugitive emissions in gas stations—elevated CH_4 levels (up to 14.1 ppm) were observed in a CNG vehicle gas station in Irvine, California (Hopkins et al. 2016).

S3 is the only site for which no good correlation was found between the mean monthly atmospheric CH_4 concentration and ambient temperature and between atmospheric CH_4 concentration and monthly consumption of NG by any of the sectors. Chen et al. (2018) showed a relatively weak seasonality of CH_4 emissions in the waste sector. However, the increase in atmospheric CH_4 in the summer, compared with the winter and spring, seems to indicate a greater participation of biogenic sources within the WWTP (Kong et al. 2002). CH_4 emissions tend to be high in summer, when biological production of CH_4 increases due to relatively high water temperature (Masuda et al. 2015). The seasonal maximum in the fall could be attributed to the predominance of non biogenic sources, associated to NG use (Sánchez et al. 2018; Wong et al. 2016) or to a greater accumulation of the CH_4 emitted by the WWTP resulting from possible thermal inversions (Verhulst et al. 2017). The latter may result from concurrent meteorological conditions (high humidity, reduced visibility, fog, low temperatures and wind intensity) (Ackerman and Knox 2006), which occur in Tandil in the fall (Picone 2014; Fusé et al. 2019). The dissimilar seasonal behavior and the relatively high atmospheric CH_4 concentrations observed in S3 required that the following analyses be performed in two ways: including and excluding S3.

All these results appear to indicate the presence of one or more dominant sources that cause atmospheric CH_4 concentrations in each site to vary across seasons. When considering the effect of ambient temperature on the mean monthly CH_4 concentration (average of concentrations in the 10 sites), a good inverse correlation is obtained ($R = -0.79$, $p = 0.002$). Therefore, the use of NG seems to account for a large percentage of atmospheric CH_4 in the urban area of Tandil, as was observed for a previous period (Fusé et.al. 2019). In turn, in Helfter et al. (2016), average CH_4 fluxes in London were 17 % lower in summer than in winter, but the correlation with air temperature was not statistically significant. This suggests that the total CH_4 flux is due to a superposition of sources with constant and time-varying emission rates.

Spatial analysis and exploratory regression of atmospheric CH_4 concentrations

Mean seasonal atmospheric CH_4 concentrations

Based on the temporal variation of the atmospheric CH_4 concentrations observed in each site, we explored differences between the sites for each season. According to the results of Fisher's LSD test, in spring, summer, and fall, atmospheric CH_4 concentrations can be classified in only two categories: high (letter *b*) for S3, and low (letter *a*) for the rest of the sites. In winter, although S3 still presents the highest CH_4 concentration, the concentrations measured in the other sites increase and the differences between the sites become more visible as a consequence (letters *a*, *b*, *c* and *d* in the results of Fisher's LSD test). This is in agreement with the results reported in Tables 1 and 2, which indicated a greater relevance of non biogenic sources associated with NG consumption during the coldest months. When repeating this analysis, excluding S3, the differences in atmospheric CH_4 concentrations between sites for each season become more evident. S8 presented the highest atmospheric CH_4 concentrations with statistically significant differences with respect to the other sites in spring and summer, and compared with S7 and

S10 in the fall. In winter, the greatest atmospheric CH_4 concentrations were measured in S6, with statistically significant differences compared with S4, S5, S7, S9, and S10.

The spatial variation in the atmospheric CH_4 concentrations measured in the city depends on the type of dominant source (fixed, diffuse, biogenic, or non biogenic), its relative contribution, and its distance from the sampling site (Carranza et al. 2018; Helfter et al. 2016). When performing the exploratory regression analysis on the 10 sites, no variable met all the search criteria established in section *Exploratory Regression* for each diagnostic test. However, some findings are worth noting. When performing it on 9 sites (excluding S3), the seasonal behavior of the sources that account for spatiotemporal variation becomes more notable (Table 3).

Table 3 Results of the exploratory regression analysis for the 9 study sites (excluding S3), by season and for the complete study period (annual): Homes connected to the gas network (GN); distance from CNG stations (GD), distance from the wastewater treatment plant (PD), distance from the artificial lake (LD)

	Variable	Sign	p-value	Adj R^2	% Signif	Passing Models
Winter	LD					
	GD	-	***	0.59	37.5	yes
	PD	-	**	0.42	12.5	
	GN	+	**	0.30		
Spring	LD					
	GD	-	*	0.28		
	PD	-		0.03		
	GN	+	**	0.55	37.5	yes
Summer	LD					
	GD	-	**	0.53	25.0	
	PD	-		0.14		
	GN	+	***	0.77	75.0	yes
Fall	LD				12.5	
	GD	-	***	0.70	37.5	yes
	PD	-	*	0.26	12.5	
	GN	+	***	0.80	62.5	yes
Annual	LD					
	GD	-	***	0.65	37.5	yes
	PD	-	*	0.29		
	GN	+	***	0.66	37.5	yes

* = 0.10, ** = 0.05, *** = 0.01

The independent variable GD met the search criteria of each diagnostic test in the fall and winter, whereas GN in the spring and summer, as well as the fall (Table 3). Still, both variables significantly correlated with atmospheric CH_4 concentrations with a value of $p < 0.05$ or $p < 0.10$ in those seasons when they failed to meet the search criteria. These results suggest that one source predominates over another according to the season. In winter, GD is the independent variable that best explains atmospheric CH_4 concentrations, while in the spring and summer, the independent variable GN accounts for them. In the fall, both of these independent variables explain the spatial variations in atmospheric CH_4 concentrations. The difference in

the predominance of the sources for each season was more easily observed in the case of residential and commercial consumption of NG than in the CNG sector (Fig. 3). This behavior was reflected in Pearson correlations between atmospheric CH_4 measured in each site and the general demand on NG for each use (Table 2); whereas in some sites residential or commercial demand on NG best explains the temporal variation in atmospheric CH_4 concentrations, in other sites it is the demand on the CNG sector the variable that accounts for them.

The variable PD did not meet the search criteria for each diagnostic test in any season. When considering the 10 sites for the exploratory regression analysis, PD presented the highest values of Adj R^2 (between 0.29 and 0.37) in the four seasons. However, PD significantly correlated with atmospheric CH_4 (Adj R^2 = 0.37 and $p < 0.10$) in 25 % of the cases reported in winter. Besides, for the linear regressions of the seasonal means for the 9 sites, the Adj R^2 value for PD was higher only in winter, with a 0.42 value but only in 12.5 % of the cases reported. The results suggest that PD alone accounts for the high atmospheric CH_4 concentrations registered in S3 whereas it only partly explains the concentrations measured in the other sites (where other CH_4 sources predominate), as the WWTP is close to only 3 sites of the sampling network (S2, S5 and S6). According to the results of the exploratory regression analysis, multicollinearity were not observed for GD and PD or GN and PD (VIF < 7.5); however, the results of the Pearson correlation test between mean monthly atmospheric CH_4 and monthly consumption of NG by sector suggest that GD or GN probably best accounts for the high CH_4 concentrations measured in these sites. This is reasonably expected especially in winter when the sources dependent on NG are the most relevant and the differences of atmospheric CH_4 concentrations between the sites become smaller.

Annual mean atmospheric CH_4 concentrations

The annual mean atmospheric CH_4 concentration measured in S3 showed statistically significant differences compared with the other sites ($p < 0.05$). The second highest value, found in S8 (zone near a CNG station and with a high density of homes connected to the gas network) (Fig. 2), only presented statistically significant differences in relation to S3 and S10. The remaining sites showed intermediate annual mean atmospheric CH_4 concentrations with no statistically significant differences between them ($p > 0.05$). The differences in atmospheric CH_4 concentrations between sites become more evident when excluding S3 from the Fisher's LSD test. S8 now presented statistically significant differences with respect to all the sites ($p < 0.05$), except S6. The rest of the sites presented intermediate annual mean atmospheric CH_4 concentrations, although with minor differences between them (represented by the pairs of letters *ab*, *bc*, *cd*, *de* of Fisher's LSD test).

Because of these observations, the exploratory regression analysis performed for the seasonal mean atmospheric CH_4 concentrations was repeated for the annual mean concentrations in order to explain the general CH_4 behavior in the city of Tandil. Once again, when considering the 10 sites for the exploratory regression analysis, no variable met the search criteria of each diagnostic test. However, some findings

are worth noting. When performing it on 9 sites (excluding S3), the variables GD and GN satisfied those criteria (Table 3).

Atmospheric CH_4 concentration correlated positively with GN (Adj $R^2 = 0.66$, $p < 0.01$) and negatively with GD (Adj $R^2 = 0.65$, $p < 0.01$). In Liu et al. (2019), population density had a remarkably positive correlation with CH_4, with a correlation coefficient of 0.74 ($p < 0.01$). As Sailor and Lu (2004) suggest, the anthropogenic heating profiles for the urban core would be correspondingly higher as they scale with population density. In Florence (Italy), road traffic and domestic heating were responsible for only 14% of the observed CH_4 fluxes, while the major residual part was likely dominated by gas network leakages (Gioli et al. 2012). A study on the megacity of Seoul, Korea, confirmed the impacts of fugitive emissions on near-surface CH_4 concentrations after the implementation of NG-powered vehicles (Nguyen et al. 2010).

PD explains 29% of the spatial variability of the atmospheric CH_4 concentration in the city with a significance $p < 0.10$. Similar results were reported by Liu et al. (2019), who found a negative correlation between PD and the atmospheric CH_4 concentration with $R^2 = 0.27$ and $p < 0.10$. When comparing both exploratory regressions for annual mean concentration, 10 sites vs. 9 sites, PD carried more weight in the first case (Adj $R^2 = 0.32$, although with $p > 0.10$ and 25% of significance of the variable). The low values of R^2 and % of significance of the variable are explained by the precise location of the source. Although both the WWTP and the CNG stations are fixed sources, these latter are located in different sites of Tandil city (Fig. 2b). For this reason, the CNG stations could generally contribute to the spatial variation in the atmospheric CH_4 concentration in the city.

No significant correlations were established with LD ($p > 0.1$); this variable failed all the tests (Table 3), proving not significant in this study. Although the urban lake was expected to acquire relevancy in the warmer months for being a biogenic CH_4 source (Ortiz-Llorente and Alvarez-Cobelas 2012), its contribution was almost nonexistent because of its fixed location in an urbanized zone.

Methane concentration associated with natural gas sources

From the results of the exploratory regression, it can be observed that when 9 sites were considered, only the models with just one variable (GN and GD) were able to meet all the search criteria established in the diagnostic tests. The reason for this may be that one of these variables could best explain the temporal CH_4 behavior in one site and, at the same time, have less relative weight in another site. For instance, the significance of GN and GD excluding S3 in the exploratory regression analysis for the entire study period was equal to 37.5 % for both variables (Table 3). This accords with the results of the Pearson correlation test between mean monthly atmospheric CH_4 in each site and monthly consumption of NG by each sector (residential, commercial, and CNG). The results reported in Liu et al. (2019) showed that the larger the population in the urban area, the greater the household energy consumption and the higher the CH_4 emissions. Particularly, as observed by Hopkins et al. (2016), high levels of atmospheric CH_4 were found

near CNG storage tanks and connecting pipes in Orange County, California; however, CH_4 increase was highly variable across the 12 different CNG stations surveyed, suggesting that fugitive leaks are responsible for these high concentrations.

In order to find a multiple OLS model that allows quantifying the interrelationships between both sources associated with NG consumption (GN and GD) and atmospheric CH_4 concentration in the city of Tandil, it would be important to incorporate more sampling sites (Quinn and Keough 2002). These should be located not only within the urban core but also towards the periphery of the city to obtain a more precise atmospheric CH_4 concentration for the entire city by increasing the measurement sites. Population density in urban cores is usually one order of magnitude higher than for the city as a whole (Liu et al. 2019). Furthermore, according to Marcotullio et al. (2013), the higher levels of CH_4 emissions per capita in urban areas, compared with non-urban areas, are due to better energy and transportation infrastructure. Consequently, mean atmospheric CH_4 concentrations and estimations per capita emissions may decrease when extending the sampling network toward the periphery. Nevertheless, it is necessary to consider that although the residential CH_4 emissions are individually small, when taken together, the sector could contribute significantly to large-scale emissions (Saint-Vincent and Pekney 2019).

Estimating the relative CH_4 contribution of sources associated with gas natural consumption, where several emission sources coexist, can be challenging. The spatial variation in atmospheric CH_4 concentration will be the result of the combined effects of various relevant factors, such as traffic variables, population or address density, land use, altitude and topography, meteorology and location, as other authors suggest (Hoek et al. 2008; Liu et al. 2019; Nisbet et al. 2020). Data availability and the unique characteristics to each area will determine the choice of variables to be used in each study, so the inclusion of a CH_4 source can be relevant in one city but not in another. In particular, the location of the WWTP in Tandil is in a densely populated area that deserves particular focus. A greater number of sampling sites around the WWTP would surely explain the spatial variability of atmospheric CH_4 concentrations around this source. In addition, extending the sampling network to the periphery of the city would entail incorporating other fixed CH_4 sources not included in this work (two WWTP and a landfill).

Conclusion

To explain spatial and temporal variations in the atmospheric CH_4 concentrations in an urban site, it is important to learn about the predominance of the sources of this gas and about the nature of each source—whether it is biogenic or non biogenic, fixed or diffuse. Identifying the main sources responsible for the increase in atmospheric CH_4 concentrations in a city helps to develop mitigation strategies. Although the WWTP is an important CH_4 source, because it is a fixed source, it only explained 29% of the spatial variation of the annual mean atmospheric CH_4 concentrations and the maximums registered during the summer. The variables “distance from CNG stations” and “number of homes connected to the gas network” are the ones that best explained the spatial variability of the annual mean atmospheric CH_4

concentrations (65% and 66%, respectively) in the urban core of the city of Tandil and the maximum concentrations registered during the fall and /or winter.

Although the sources associated with NG consumption (residential, commercial, and CNG) cause only minor increases in CH_4 concentrations, they are scattered in the whole urban area, and together their relative contribution therefore increases. Improving the efficiency of each system involved, from NG distribution, residential and commercial consumption to CNG use in vehicles, would not only reduce the emissions of this potent greenhouse gas and its resulting impact on the environment, but would also reduce gas losses and consequently bring economic benefits to each of these sectors. Preventing NG leaks to reduce emissions in all urban areas should be a goal to achieve in the short term.

Based on the results shown here, we propose expanding the sampling network and also performing the same study adjusting the model using the same and other variables. Finally, the baseline atmospheric CH_4 concentration for the urban area obtained in this study, would allow estimating how the city growth would contribute to atmospheric CH_4 as a result of the relative increase of the number of sources in the periphery of the city. In this sense, the results obtained serve as an important reference for medium-sized cities in constant growth.

Declarations

Ethics approval and consent to participate

Not applicable

Consent for publication

Not applicable

Availability of data and materials

The datasets generated during the current study are available in the Mendeley Data repository: https://data.mendeley.com/datasets/c5n4k6ddw7/1. Fusé VS, Stadler CS, Picone N, Linares S, Guzmán SA, Juliarena MP (2021) Atmospheric methane concentrations in the urban core of a medium-sized city. Mendeley Data V1. https://doi.org/10.17632/c5n4k6ddw7.1

Competing interests

The authors declare that there is no conflict of interest.

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Authors' contributions

Investigation and Methodology were performed by VSF. Formal analysis and Visualization were performed by VSF and CSS. Conceptualization was performed by VSF, SL and MPJ. SAG and MPJ provided the necessary resources to carry out the study. NP provided the necessary ArGIS resources to explain the spatial variability of the mean methane values obtained. Funding acquisition was performed by SL and MPJ. MPJ was responsible for project administration and Supervision. The first draft of the manuscript was written by VSF and all authors commented on previous versions of the manuscript. All authors read and approved the final manuscript.

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Figures

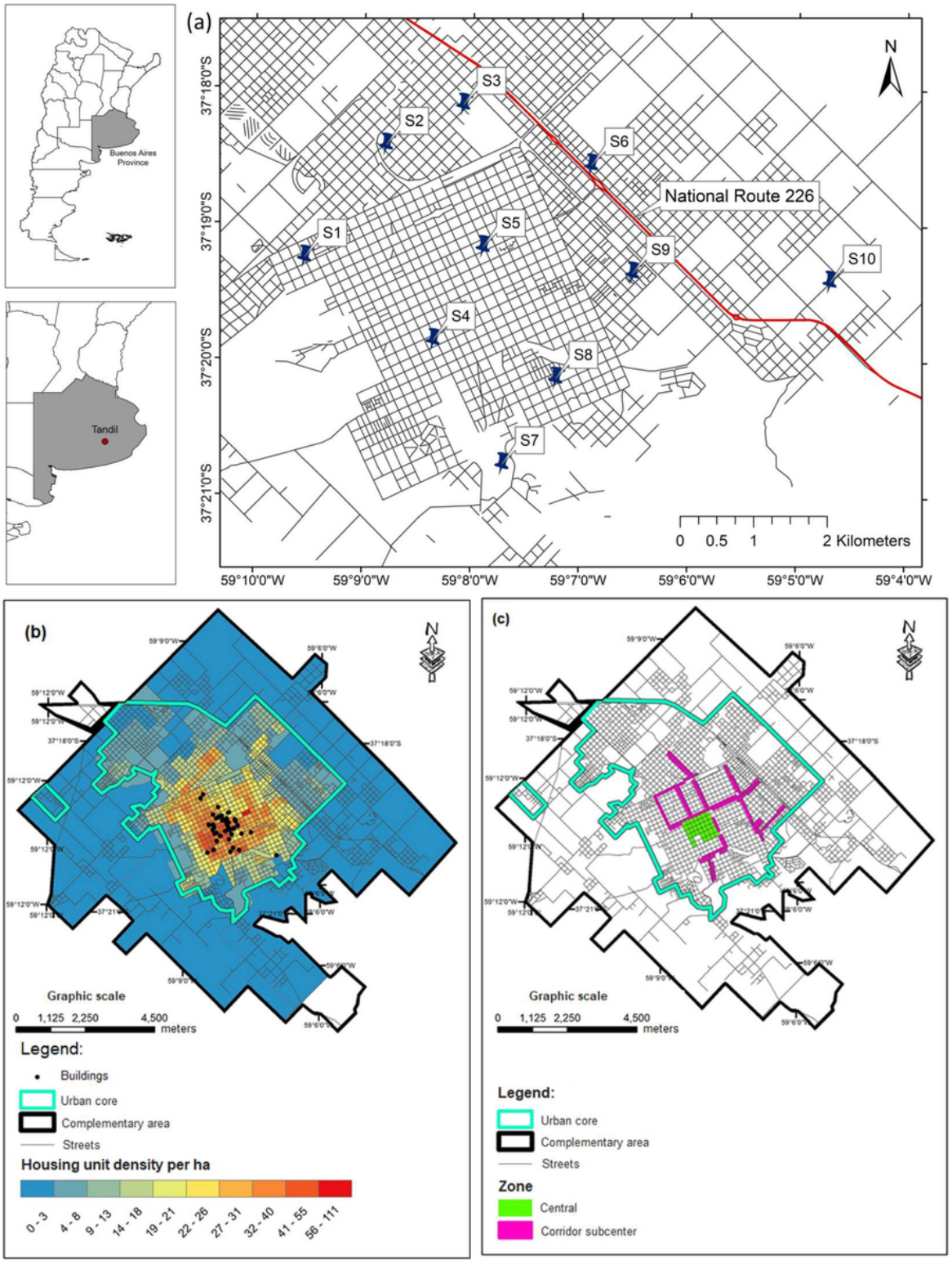

Figure 1

a Geographical location of the sampling sites; b residential areas and c commercial areas in Tandil city [Migueltorena and Linares 2019]

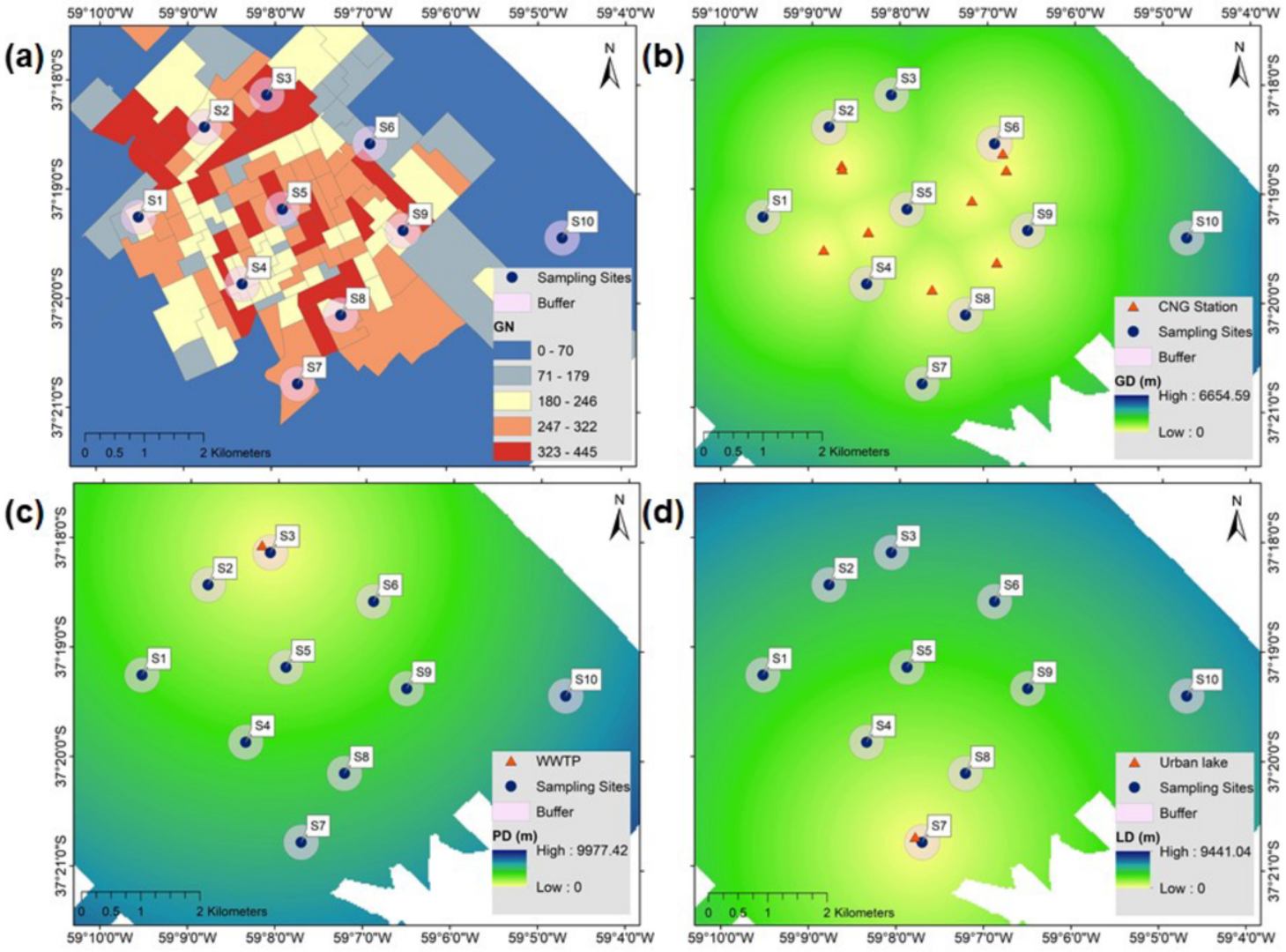

Figure 2

Sampling sites and Geographical Information System (GIS) layers: a number of homes that use gas for cooking within census tract (natural gas network, GN) (INDEC 2010); b distance from CNG station (GD); c distance from wastewater treatment plant (PD); d distance from the urban lake (LD)

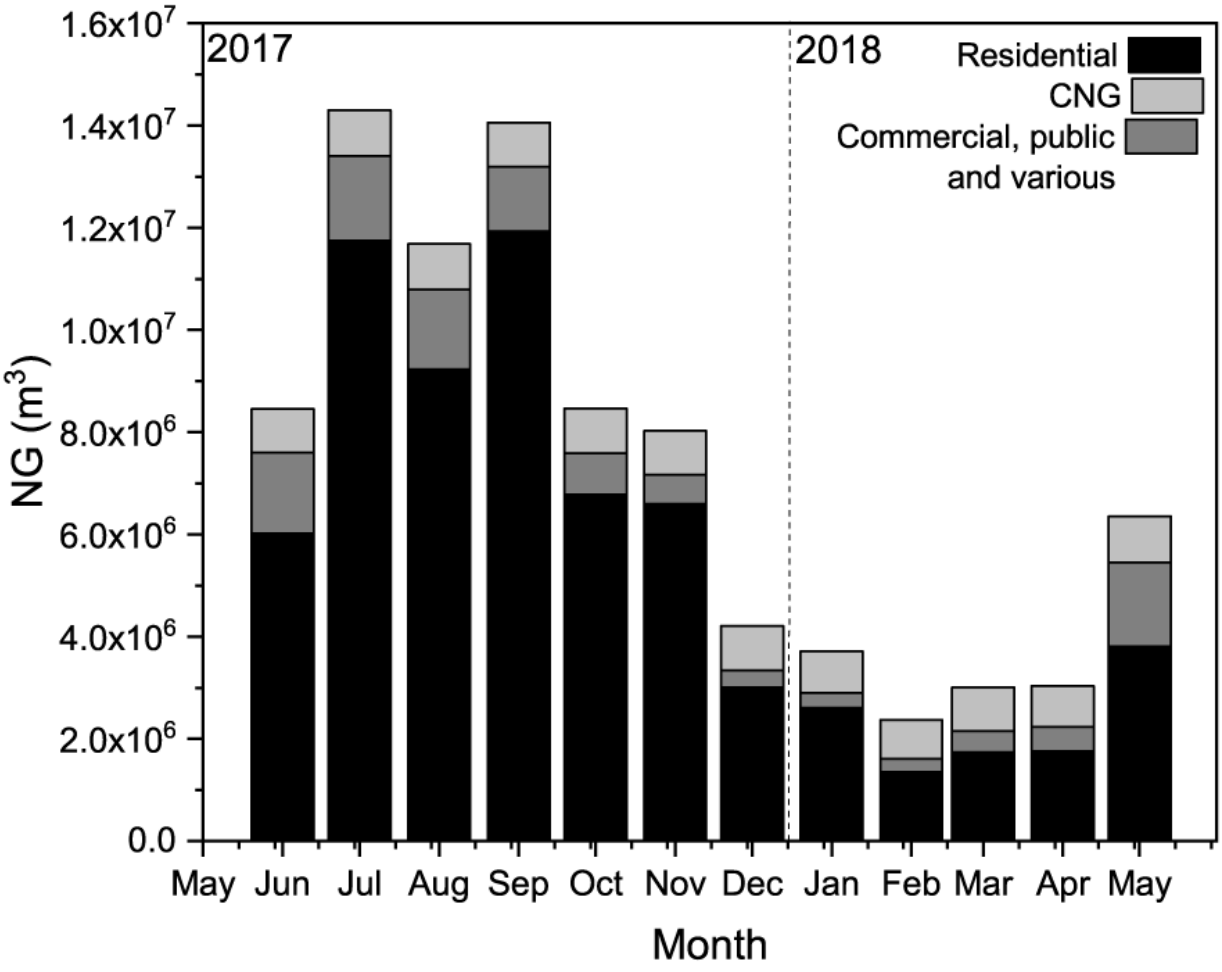

Figure 3

Natural gas (NG) consumption in cubic meters (m3) for each sector (residential, compressed NG [CNG], commercial, public, and various)