Source apportionment vs. emission inventories of non-methane hydrocarbons (NMHC) in an urban area of the Middle East: local and global perspectives

T. Salameh[1,2,3*], S. Sauvage [1,2], C. Afif [3], A. Borbon [4], and N. Locoge [1,2]

[1] Mines Douai, Sciences de l'Atmosphère et Génie de l'Environnement (SAGE), F-59508 Douai Cedex, France
[2] Université de Lille, 59000 Lille, France
[3] Emissions, Measurements, and Modeling of the Atmosphere (EMMA) Laboratory, Unité Environnement, Génomique Fonctionnelle et Études Mathématiques, Centre d'Analyses et de Recherche, Faculty of Sciences, Saint Joseph University, Beirut, Lebanon
[4] Laboratoire Interuniversitaire des Systèmes Atmosphériques (LISA), IPSL, CNRS, UMR 7583, University of Paris Est Créteil (UPEC) and Paris Diderot (UPD), Créteil, France
* : Now at Laboratoire Interuniversitaire des Systèmes Atmosphériques (LISA), IPSL, CNRS, UMR 7583, University of Paris Est Créteil (UPEC) and Paris Diderot (UPD), Créteil, France
Therese.salameh@lisa.u-pec.fr

Abstract:

We applied the Positive Matrix Factorization model to two large datasets collected during two intensive measurement campaigns (summer 2011 and winter 2012) at a sub-urban site in Beirut, Lebanon, in order to identify NMHC sources and quantify their contribution to ambient levels. Six factors were identified in winter and five factors in summer. PMF-resolved source profiles were consistent with source profiles established by near-field measurements. The major sources were traffic-related emissions (combustion and gasoline evaporation) in winter and in summer accounting for 51 wt. % and 74 wt. % respectively in agreement with the national emission inventory. The gasoline evaporation related to traffic source had a significant contribution regardless of the season (22 wt. % in winter and 30 wt.% in summer). The NMHC emissions from road transport are estimated from observations and PMF results, and compared to local and global emission inventories. The PMF analysis finds reasonable differences on emission rates, of 20 – 39 % higher than the national road transport inventory. However, global inventories (ACCMIP, EDGAR, MACCity) underestimate the emissions up to a factor of 10 for the transportation sector. When combining emission inventory to our results, there is a strong evidence that control measures in Lebanon should be targeted on mitigating the NMHC emissions from the traffic-related sources. From a global perspective, an assessment of VOC anthropogenic emission inventories for the Middle East region as a whole seems necessary as these emissions could be much higher than expected at least from the road transport sector.

Keywords: VOC; Positive Matrix Factorization (PMF); urban area; emissions; traffic; emission inventories

Highlights:

- PMF model was applied to identify major NMHC sources and their seasonal variation.
- Gasoline evaporation accounts for more than 40% both in winter and in summer.
- NMHC urban emissions are dominated by traffic related sources in both seasons.
- Agreement with the national emission inventory regarding the relative contribution of the on-road mobile source.
- Disagreement in terms of emission quantities suggesting an underestimation of the global inventories.

1. Introduction

Fast urbanization expansion, high population density, industrialization, intensive transport networks, and negligent pollution control have led to a substantial degradation of air quality in the Middle East (MEA) region (UNEP, 2006). Nowadays, air quality is taken as a major aspect of the quality of life leading to sustainable development in many areas of the world.

In Lebanon, a developing country in the MEA, which is located in Western Asia on the eastern shore of the Mediterranean Sea, air quality has been continuously deteriorated leading to high pollutant levels exceeding the World Health Organization (WHO) recommended values (WHO, 2005); for instance, Afif et al. (2009) reported an annual average concentration of nitrogen dioxide (NO_2) in Beirut from December 2004 to June 2006, of 67 µg/m^3 which is higher than the WHO annual recommended value of 40 µg/m^3 (WHO, 2005). In addition, high levels of particulate matter, PM_{10} and $PM_{2.5}$, were obtained with annual concentrations of 64 and 20 µg/m^3 respectively (Massoud et al., 2011) exceeding WHO guideline values of 20 and 10 µg/m^3. Although these measurements provide valuable information on air pollution, they are scarce and limited to a few pollutants. In the absence of adequate environmental policy and action, Lebanon is facing today significant urban air pollution problems; the cost of environmental degradation of air quality was last estimated in 2001 at 1.02% of the annual gross domestic product (GDP) (Sarraf et al., 2004).

Among air pollutants, non-methane hydrocarbons (NMHCs) play an important role in urban areas since they contribute to the formation of photochemical oxidants such as ozone and peroxyacetylnitrate (PAN) and they are also involved in the formation of secondary organic aerosols (SOA) (Seinfeld and Pandis, 2006). Moreover, some species are associated with adverse health impact or even are carcinogenic (e.g. benzene, 1,3- butadiene, etc.) (WHO, 2000).

NMHCs are emitted mainly from anthropogenic sources in urban areas. Major anthropogenic sources include vehicle exhaust, fuel evaporation, solvent use, emissions of natural gas and industrial processes (Friedrich and Obermeier, 1999, Sauvage et al., 2009). While urban emission sources of NMHC are usually well identified, their relative importance is still under debate and can be region-dependent (Yuan et al., 2013).

Considering the diversity in chemical reactivity among NMHCs, their different ozone and SOA formation potentials (Carter 1994, Derwent et al., 2010) and the speciation profiles of the various emission sources, there is a clear need to identify and quantify NMHC emission sources, and their associated speciation by the development of regional emission inventories in order to apply efficient control strategies and air pollution management.

Moreover, emission inventories are used as input data for air quality models; therefore, their accuracy is also important for air quality impact assessment. However, recent in-situ observations have revealed persistent and high uncertainties in emission inventories in urban areas of post-industrialized countries. Borbon et al. (2013) found large discrepancies between observed emissions ratios and those calculated from the inventories in Los Angeles and Paris. Moreover, Gaimoz et al. (2011) showed that the current emission inventory for Paris strongly overestimates the volatile organic compounds (VOCs) emitted from solvent use and needs to be corrected. Niedojadlo et al. (2007) also revealed some inconsistency between emission inventory in Germany and source apportionment results, especially regarding emissions related to solvent use. Those studies have revealed that source-receptor approaches could be a relevant alternative to emission inventories where provided observations are available.

For the MEA region, emissions uncertainties could be even higher. At present, local inventory/emission data and observations for their evaluation are sparse and not speciated, potentially resulting in higher uncertainty in modeling approaches (Waked and Afif, 2012, Waked et al., 2013). In particular, there is a paucity of data on NMHC levels and their emission sources in the Middle East region. At the global scale, several emission inventories exist but very few of them are speciated or exhibit fine temporal resolution (ECCAD database). For Lebanon, a recent work has led to the development of an atmospheric emission inventory of anthropogenic and biogenic sources considering CO, NO_x, SO_2, NMVOC, NH_3, PM_{10}, and $PM_{2.5}$, for a base year of 2010 according to the 2009 EMEP/EEA guidelines (Waked et al., 2012).

The aim of this work is to identify major NMHC sources and quantify their contribution to NMHC concentrations on a seasonal basis by applying the source-receptor Positive Matrix Factorization model (PMF) to the observations collected at a suburban site in the urban area of Beirut, Lebanon, and evaluate its consistency with existing emission inventories. The receptor modeling techniques are numerous; many of them have been previously used in NMHC source apportionment worldwide including Principal Component Analysis (PCA), Positive Matrix Factorization (PMF), Chemical Mass Balance (CMB) and UNMIX like in Europe (Badol et al. 2008, Sauvage et al. 2009), the USA (Leuchner and Rappenglück 2010), and in China (Guo et al. 2007, Yuan et al. 2013). The performance of four receptor models (PCA, PMF, CMB and UNMIX) was evaluated by Miller et al. (2002) by applying them to the same artificial dataset of VOCs. They found that PMF extracted factor profiles that most closely represented the major sources used to generate the simulated data, and concluded that PMF is the most appropriate model to explain the results (Miller et al., 2002).

A large set of speciated NMHC have been continuously measured during two-week periods in summer 2011 and winter 2012 in the frame of the Emission and Chemistry of Organic Carbon in the East Mediterranean (ECOCEM) experiment. The consistency of the source composition obtained by the PMF on a seasonal basis is evaluated with regards to NMHC source profiles established by canister sampling in the vicinity of emission sources (Salameh et al., 2014). Finally, PMF results are used to evaluate local and global emission inventories.

2. Experimental procedures

2.1. Sampling site: location and surroundings

The site was chosen in a way to be far enough from strong direct emissions (industrial, road transport) since they can hide the emissions from distant sources. At the same time, the site is appropriately located in order to receive the air masses coming from Greater Beirut Area (Figure 1). The measurements were conducted on the roof of the Faculty of Sciences building of Saint Joseph University (33°87' N, 35°56' E) from 2 to 18 July 2011 in summer and again from 28 January to 12 February 2012 in winter. The site is located in the eastern suburb of the city of Beirut (6 km South-East of Beirut downtown) at an altitude of 230 m ASL (Above Sea Level). The site is surrounded by a forested pine area and a high density of residential premises. Beirut International Airport is located 8 km southwest of the site and one fuel storage facility is located at 3.6 km north (Salameh et al., 2014).

2.2. Material and methods

NMHCs were continuously analyzed by on-line Thermal Desorption Gas Chromatography with a Flame Ionization Detector (TD-GC-FID) provided by Perkin Elmer described elsewhere (Salameh et al., 2014). The on-line measurement was performed hourly, covering 30 minutes of ambient air sampling. 67 NMHCs from C2 to C9 belonging to alkanes (29), alkenes (19), alkynes (2) and aromatics (17) were identified and quantified. The detection limits were around 40 pptv for the targeted compounds except for ethane and ethylene (90 pptv). A certified NPL (National Physical Laboratory) standard of NMHC mixture (~4 ppbv) was used to determine and check the stability of the GC carbon response during both measurement periods.

Additional trace gases including carbon monoxide (CO), NO_x and O_3 were measured on-line on a 1 min basis using trace gas analyzers. Basic meteorological parameters (wind speed and direction, temperature, relative humidity and atmospheric pressure) were measured on a 1 min basis during both campaigns.

3. Source apportionment by Positive Matrix Factorization (PMF)

3.1. PMF model description:

The USEPA PMF v3.0 was applied for NMHC source apportionment analysis. This method is described in details elsewhere (Paatero and Tapper, 1994, Paatero, 1997). NMHCs which are not identified during both seasons (like tetrachloroethylene) or their concentrations were frequently below the detection limits, were not considered in the PMF analysis.

According to the PMF, an observed concentration at a receptor site can be viewed as a data matrix X of i by j dimensions in which i number of samples and j chemical species were measured.

$$x_{ij} = \sum_{k=1}^{p} g_{ik} f_{kj} + e_{ij} \tag{1}$$

where g_{ik} is the mass contribution of the k_{th} source to the i_{th} sample, f_{kj} is the j_{th} species mass fraction from the k_{th} source, p is the number of independent sources, and e_{ij} is the residual associated with the concentration of the j_{th} species in the i_{th} sample. The solution of the equation is obtained by minimizing the residual sum of squares Q given by:

$$Q = \sum_{i=1}^{n}\sum_{j=1}^{m}\frac{e_{ij}^2}{s_{ij}^2} = \sum_{i=1}^{n}\sum_{j=1}^{m}\left(\frac{x_{ij} - \sum_{k=1}^{p} g_{ik} f_{kj}}{s_{ij}}\right)^2 \tag{2}$$

where $f_{kj} \geq 0$ and $g_{ik} \geq 0$ and where n is the number of samples, m the number of considered species, and s_{ij} an uncertainty estimate for the j_{th} species measured in the i_{th} sample.

The results are constrained so that no sample can have significantly negative source contributions. The goal is to identify the number of factors p, the species profile f of each source, and the amount of mass g contributed by each factor to each individual sample by using measured source profile information, meteorological parameters analysis, and additional species.

3.2. PMF model implementation:

One of the advantages of the PMF is to consider the uncertainty of each individual concentration. Special attention was paid to the estimation of the uncertainty. For each measured concentration, an uncertainty was calculated following the method developed within the ACTRIS (Aerosols, Clouds, and Trace gases Research InfraStructure) network (Hoerger et al., 2015, see supplemental material). It includes the systematic errors in the measurement and the precision which reflects the random errors. Thus, the total uncertainty of a measurement is obtained with equation 3:

$$\Delta x^2_{\text{unc}} = \Delta x^2_{\text{precision}} + \Delta x^2_{\text{systematic}} \tag{3}$$

The precision is derived from a series of measurements of the NPL standard gas. It covers the random errors of the sampling and analytical system.

Possible systematic errors include the calibration gas uncertainty, systematic peak integration errors, sample volume determination, and potential blank value (Hoerger et al., 2015, see supplemental material). Therefore, the overall systematic error is described as follows:

$$\Delta x^2_{\text{systematic}} = \Delta x^2_{\text{calibration}} + \Delta x^2_{\text{integration}} + \Delta x^2_{\text{volume}} + \Delta x^2_{\text{blank}} \tag{4}$$

The systematic integration error can be neglected because the chromatograms of the air samples and the calibration gas for the considered species were quite similar, without any integration issues.

The systematic volume error is neglected since $\delta V_{\text{sample}} = \delta V_{\text{calibration}}$.

For the expanded uncertainty, the total uncertainty calculated is multiplied by the coverage factor k=2.

The relative expanded uncertainty ranged between 4 and 27% in summer for all the compounds excepting for octane which was 38%. In winter, the relative expanded uncertainty was between 5 and 35% for most of the compounds excepting for trimethylbenzenes, isoprene, octane and heptane which had an uncertainty between 40 and 60 %.

The missing data were replaced by the median of the concentrations measured the other days at the same hour; and in these cases, the uncertainty was set as four times the concentration. Values reported as below DL were replaced by half of the DL and the corresponding uncertainty was set as 5/6 of the DL value.

Paatero and Hopke (2003) have introduced the signal-to-noise ratio (S/N) which indicates whether the variability in the measurements is real or within the noise of the data. If the S/N ratio is less than 0.2, the species is excluded and if the ratio is greater than 0.2 but less than 2, the uncertainty is multiplied by four.

For a reliable identification of the sources, the PMF requires a large number of samples. The large datasets collected during the two campaigns were lumped separately for PMF source apportionment seasonal analysis. By taking into consideration the different existing reactivities, mainly with OH radical within the NMHC species, the profile of each source may change temporally during transport of emission plumes. Nevertheless, we have examined the factors influencing the distribution of NMHCs (Salameh et al., 2015) and we concluded that NMHCs were freshly emitted from local sources in winter but also in summer, and the chemical losses of NMHC species, even for the most reactive species, were likely not significant.

3.3. Determination of the optimal solution:

Several base runs were performed with different number of factors from 3 to 12. The diagnostic parameters include the value of Q, IM which is the maximum individual column mean and IS which is the maximum individual column standard deviation defined by Lee et al. (1999). Q, IM, and IS are then plotted against the number of factors (from 3 to 12). The number of factors chosen corresponds to a significant decrease of Q, IM, and IS. In order to control the rotation and to optimize the selected solution, the F_{peak} parameter was used. Six factors were extracted from winter dataset and five from summer dataset. Quality indicators from the PMF application are summarized in Table 1.

4. Results and discussion

4.1. Meteorological conditions

Lebanon is characterized by a narrow coastal strip located on his western part and by the Bekaa valley located between the Western and Eastern Lebanese mountain chains. The coastal region has a Mediterranean climate with land–sea breeze circulation. During the summer measurement campaign, the temperature at the Saint Joseph University site ranged from 20 to 29°C with an average of 25°C. The average wind speed was 2 $m.s^{-1}$ with a maximum wind speed of 10 $m.s^{-1}$ during the day coming mostly from the South-West and from the North on July 7, 8 and 9. The wind direction was mostly from the North-East and slightly from South, South-West at night. During the winter measurement campaign, the temperature ranged from 7 to 22°C with an average of 13°C. The average wind speed was 2 $m.s^{-1}$ and the wind direction was mostly coming South-Easterly and Easterly (Salameh et al., 2015).

4.2. Identification of PMF profiles

In order to attribute PMF factors to emission sources, a comparison between extracted PMF profiles and speciated profiles established in the vicinity of emission sources by Salameh et al. (2014) was carried out, when possible. In addition, the diurnal cycle and time series of the contributions of modeled sources were analyzed with independent parameters like meteorological data and specific tracers like CO and NO_x.

4.2.1. Winter PMF factor identification and contribution

In winter, six sources were identified corresponding to combustion mainly related to regional traffic, combustion related to local traffic, gas leakage, gasoline evaporation related to traffic, gasoline evaporation as an episodic point source and a source characterizing the urban background. The composition of the source profiles and the diurnal variations of the source contribution are reported respectively in Figures 2 and 3.

Combustion mainly related to regional traffic

Factor 1 has significant amounts of toluene, m,p-xylenes, ethylbenzene, ethylene, acetylene, 1,2,4-trimethylbenzene & decane (co-eluted), and benzene which are typical combustion products. Almost 30% of the variances of ethylene, propene, acetylene and benzene are explained by this factor. This profile is very similar to the profile determined in the vicinity of the emission sources in Beirut which consists of exhaust and evaporative running losses from vehicles (Salameh et al., 2014). The factor contribution is well correlated with the concentrations of CO and NO_x which are combustion tracers (R=0.8 and 0.7 respectively). The daily time series of the contribution of this source are characterized by a first significant increase of concentrations at around 07:00-08:00 LT (UTC/GMT+2 h), the second one at 14:00-15:00 LT, another increase at 17:00-18:00 LT and a small peak is observed at 20:00-21:00 LT in the evening. According to Waked et al. (2012), the diurnal variation profile for on-road mobile source in urban areas in Lebanon shows two significant increases in weekdays, a morning peak at around 07:00-10:00 LT and an evening peak from 16:00 to 18:00 LT consistent with the important peaks observed for factor 1. In addition, in winter, additional sources related to domestic heating should be considered since the measurement site is surrounded by residential area. The average relative contribution of this factor is 19 % in winter.

Factor 2 is also characterized by some combustion tracers (ethylene, acetylene, toluene, benzene, propene, and m,p-xylenes). Nearly 50% of the variances of ethylene as well as propene are explained by this factor. The factor contribution strongly correlates with CO and NO_x (R=0.8 and 0.7 respectively). This factor is associated to "combustion related to local traffic" since it doesn't show a significant contribution during the weekend which is due to the limited local activity nearby the measurement site (schools, universities, residential area). The daily variation of this factor is characterized by an initial increase at 07:00-08:00 LT, a peak at 18:00-19:00 LT and the last peak in the evening at 20:00-21:00 LT. These enhancements are far less than those of factor 1. The average relative contribution of this factor is 10% in winter.

Gasoline evaporation related to traffic

Isopentane, butane, isobutane, toluene and 2-methylpentane are the main compounds of **factor 3**. Most of isopentane (53%) and around 36% of butane and isobutane are explained by this factor. This strongly suggests that this profile is related to fuel evaporation. Additionally, the profile of this factor, taking into account the larger NMHC contributors, is compared to three gasoline evaporation profiles established by near field measurements including the fuel storage facilities, hot soak (a type of evaporative emissions which occurs when a warmed-up vehicle is stationary and the engine is stopped) and refueling cars (Salameh et al., 2014) in figure 4. The fingerprint is consistent within all the gasoline evaporation source profiles confirming the nature of this source. In Lebanon, light-duty vehicles operate on gasoline while only heavy duty vehicles are allowed to run on diesel (4 % of fleet in Beirut) (Waked and Afif, 2012). The diurnal profile as well as the time series in figure 5 display the same aforementioned peaks in factors 1 and 2 especially in the morning and in the evening assuming that the gasoline evaporation is linked to traffic. The average relative contribution of this factor is 22 % in winter.

Gasoline evaporation

Factor 5 is also characterized by the C4-C5 alkanes, toluene and 2-methylpentane. Nearly 42% of the isopentane and pentane variability and 48% of the isobutane and butane variability are explained by this factor. The fingerprint of the chemical composition is consistent with the ones of typical gasoline evaporation profiles established by near field measurements (fig. 4). The diurnal profile of this source contribution is marked by a night minimum and an increase in the midday. The time series presented in figure 5 shows that this factor (F5) is primarily episodic with high contribution originating from the North wind sector as demonstrated in the pollution rose in figure 6. The extremely high levels of the source contribution come from the North sector where the fuel storage facilities are located (fig. 6). A collinearity between both gasoline evaporation factors (F3 and F5) is observed around noon when the wind blows from the North sector since the highest contributions are detected in this sector making the apportionment difficult. The average relative contribution of this factor is 21 % in winter.

Gas leakage

58% of propane variability is explained by **factor 6**. The composition of this source is mainly impacted by high loadings of propane, butane and isobutane which are the signatures of LPG (Liquified Petroleum Gas) (Blake and Rowland 1995). Hence, this source is attributed to gas leakage mainly LPG from residential use. LPG is the main fuel for cooking in households in Lebanon and it is used, among others, for domestic heating. The diurnal profile of this factor does not show significant peaks, only some small peaks are observed in the morning, at noon and in the late evening (fig. 3). The average relative contribution of this factor is 10 % in winter.

Background

Long-lived species are dominant in **factor 4**. The ethane and propane variability explained by this factor is 76% and 42% respectively, even though, toluene, butane and acetylene are also present in this profile. This factor is, therefore, categorized as urban background (Lanz et al. 2008, Sauvage et al. 2009). The diurnal profile of this factor does not show any significant peak and instead maintains the same contribution independently of the hour of the day (fig. 3). The average relative contribution of this factor is 18 % in winter.

4.2.2. Summer PMF profile identification and contribution

One of the limitations of PMF application for summer data could be the effect of the photochemistry (Yuan et al., 2012). The effect of photochemistry has been assessed in Salameh et al. (2015), and the results showed a limited influence of the photochemistry in summer.

The five sources identified by the PMF analysis in summer correspond to combustion related (so-called nocturnal and diurnal) to traffic, gasoline evaporation related to traffic, gasoline evaporation as an episodic point source and a source characterizing the local background. The composition of the source profiles and the diurnal variation of the source contribution are reported respectively in Figures 7 and 8.

Traffic related sources

Two profiles (factor 1 and factor 3) related to traffic have been extracted with different contributions between day and night.

The profile of **factor 1** is essentially composed of aromatic compounds toluene, m,p-xylenes, o-xylene, ethylbenzene, 1,2,4-trimethylbenzene & decane (co-eluted), ethylene, acetylene and isopentane which are included in the traffic related sources (gasoline evaporation and combustion, fig. 9) and are present in the composition of factor F1 in winter (especially the aromatics). The diurnal variation shows high loadings during the day (fig. 8). A significant first increase is observed at

07:00-08:00 LT (UTC/GMT+3 h), two other small peaks are observed during the afternoon (14:00-15:00 LT) and in evening (20:00-21:00 LT) comparable to the peaks observed in the combustion related to traffic during winter (winter F1). This factor is linked to traffic emissions occurring during the day and coming mainly from the dominating daytime the southwest sector. The average relative contribution of this factor, combustion related to diurnal traffic, is 17 % in summer.

Typical combustion tracers are the major contributors to the profile of the **factor 2**. The variability of acetylene (34%), ethylene (43%), propene (45%) and benzene (30%) is mostly explained by this factor. The daily variability of this factor displays high contributions during nighttime characterizing the combustion related mainly to nocturnal traffic. At night, the wind direction is mostly from the northeast, the South, and southwest where the highway network is densely occupied at night in summer and the vehicles run slowly. When the vehicles speed is low, especially in the case of vehicles running on gasoline without a functioning catalytic converter (Waked and Afif 2012), the NMVOC emissions increase (Fontaine 2000). The average relative contribution of this factor is 27 % in summer.

Gasoline evaporation related to traffic

Factor 3 encloses a high percentage of butane, toluene, propane, m,p-xylenes and isopentane which strongly suggests that this factor is related to the gasoline evaporation. This factor exhibits also high loadings during nighttime alike the combustion related to nocturnal traffic factor 2 confirming the traffic as the origin of these two factors (F2 and F3).

By comparing both combustion (F1 and F2) and gasoline evaporation (F3) factors related to traffic profiles with the road transport profile established in near-field measurements (Salameh et al., 2014), which consists of exhaust and evaporative running losses from vehicles (figure 9), we notice the same overall fingerprint for the majority of the species, nevertheless, propane exhibits a high percentage in the gasoline evaporation profile and 75% of its variability is explained by this factor (F3) suggesting the liquefied petroleum gas leakage as an additional source to the gasoline evaporation. The average relative contribution of this factor is 30 % in summer.

Gasoline evaporation (episodic point source)

Factor 5 is characterized by C4-C5 alkanes, toluene and 2-methylpentane similar to the factor 5 composition in winter. This factor is gasoline evaporation showing a high contribution in the middle of the day. The pollution rose, as in winter, shows high contributions coming from the North sector, where the fuel storage facilities are located, as shown in figure 10 which corresponds to episodic events having occurred on July 7, 8 and 9. The average relative contribution of this factor is 16 % in summer.

Local background

64% of the variability of isoprene, commonly used as the tracer of biogenic emissions, is explained by the **factor 4**. The profile of this factor consists largely of toluene, ethane, isopentane, m,p-xylenes, acetylene, butane and propene which are related to combustion. Therefore, “local background” is attributed to this factor including the regional background, the combustion from the surrounding sources, as well as the biogenic emissions from the forest nearby the measurement site. It is not possible to separate this type of source even by increasing the number of PMF factors. The diurnal profile shows higher contributions during the day starting 07:00 – 08:00 LT and lower contributions during the night. The average relative contribution of this factor is 10 % in summer.

5. Similarities and differences between summer and winter results

Figure 11 summarizes the average relative contribution in weight % of each factor in winter and in summer. We found several common factors in winter and summer but their relative contribution was slightly different. For instance, the contribution of traffic related sources including the combustion and the gasoline evaporation from traffic accounted for 51% in winter and 74% in summer. Nevertheless, the episodic gasoline evaporation source contributed to 21% in winter and 16% in summer. This source was a major contributor to winter’s NMHC concentrations reflecting the importance of gasoline evaporation in winter. If we consider the gasoline evaporation sources (episodic and related to traffic), we obtain a significant contribution of 43% in winter and 46% in summer.

Additional factors were found in winter but absent in summer. For instance, gas leakage due to the use of LPG in residential heating. Another factor characterizing the urban background highly contributed to winter's NMHC concentrations (18 %) mainly because of the meteorological conditions and the absence of photochemistry in winter (Salameh et al., 2015). A source characterizing the "local background" including the biogenic emissions was exclusively found in summer since the meteorological conditions are favorable. Nevertheless, this "local background" also includes regional background and combustion emissions.

Based on the PMF analysis, we conclude that the main contributors to the NMHCs are the combustion and gasoline evaporation sources in summer as well as in winter. These sources are of local origin confirming what has been demonstrated previously (Salameh et al., 2015) that there is no significant influence of long range transport of air masses on the NMHC concentrations.

6. Intercomparison with the National Emission Inventory for Lebanon -NEI-

The contribution of the factors discussed above is dependent on the seasons and associated meteorological conditions, therefore, care must be taken when comparing these results to the annual national emission inventory (NEI) for Lebanon.

The NEI is not speciated, and was established for a base year of 2010 within the EEA/EMEP guidelines, by Waked et al. (2012), covering major anthropogenic and biogenic sources in the region with 5 km spatial resolution for Lebanon and 1 km spatial resolution for its capital city Beirut and its suburbs. The total VOC emissions in Lebanon were estimated to be 115 Gg for the year 2010. According to this inventory, transport is the main source of NMVOCs with a relative contribution of 67% of total emissions of VOCs in Beirut and its suburbs as well as in all over Lebanon. PMF results give nearly the same contribution of this sector in winter 51% and 74% in summer. According to Waked and Afif (2012), the gasoline evaporation contribution was evaluated to be responsible for 8% of total NMVOC emissions in Beirut and its suburbs and 17 % of total NMVOC emissions at the national level based on gasoline consumption whereas the PMF gasoline evaporation (related to traffic) sources contributed to 22% in winter and 30% in summer.

The comparison held here is subject to several limitations. On one hand, the national emission inventory is not speciated, and it needs further improvements regarding the input data and the Lebanon-specific emission factors need to be established (Waked et al., 2012). On the other hand, the measurement site is qualified as sub-urban and is located far from some sources identified in the inventory, like for instance large forests representing biogenic sources, and industrial plants. These sources have not been identified in the PMF-resolved profiles since they weren't major sources during this study and some of them are located in the North and South of Lebanon far away from the measurement site. Additionally, it was hard to discern the biogenic source since the key tracers of this source such as terpenes and oxygenated volatile organic compounds (methanol, acetone…) were not included in the PMF analysis. Furthermore, the emission inventory data used for comparison are provided on annual statistical data, on a national scale whereas the data here is obtained from a single measurement site during a shorter time regarding a non-exhaustive list of species. Therefore, long term and continuous studies integrating more than one measuring site and more specific tracers are of great interest in order to provide more reliable information.

7. Comparison of PMF results and emission inventories for road transport

Our study provides the first comprehensive observational constraints for a quantitative evaluation of anthropogenic emission inventories at local, national (Waked et al., 2012) and global levels for a Middle Eastern area. While such comparison is somewhat limited as the basis for comparison is different (NMHC speciation, number of NMHCs, temporal and spatial resolution), it still provides relevant insights on uncertainties and perspectives of improvement. The objective of this last section is to estimate NMHC emissions from the road transport sector from PMF results and carbon monoxide observations and to compare these values to the ones derived from various emission inventories.

7.1. Comparison to the National Emission Inventory for Lebanon -NEI-

The annual emissions of air pollutants originating from on-road mobile sources in 2010 in Beirut specifically and in Lebanon were investigated by Waked and Afif (2012). At a national level, the estimated annual emissions for CO were 522 Gg and 48 Gg for NMVOCs by using the EMEP methodology (Waked and Afif, 2012). In parallel, we calculated separately, for each season, the ratio of the sum of the 59 NMHC contributions (in µg/m^3) coming from the PMF factors related to road transport (combustion and gasoline evaporation) and of the mean of the CO measured (in µg/m^3) during each measurement campaign according to equation (5). Then, we used the estimated annual emissions for CO (Q) in Gg to generate the emissions of the 59 NMHCs based on the winter ratio on one hand, and on the summer ratio on the other hand, in order to give a range of annual emissions levels.

$$\text{NMHC} = \text{ratio (NMHC/CO)}_{\text{ECOCEM}} \times \text{Q}_{\text{CO inventory}} \tag{5}$$

With NMHC = Sum of 59 measured species during ECOCEM (in Gg).

The mean concentration of CO was equal to 470 ± 195 µg/m^3 during summer campaign, while it was equal to 388 ± 228 µg/m^3 during winter campaign. The measured NMHC to CO ratio was estimated to 0.15 in summer, and to 0.12 in winter.

By this method, the annual emissions for Lebanon are estimated to 78 Gg from the summer ratio and to 62 Gg by the winter ratio, which are higher than the annual emissions of 48 Gg. In both cases, the emissions from the national road transport inventory are lower with a difference ranging from 22% for winter based emissions to 39% for summer based emissions (figure 12). These results suggest that the inventory underestimates the road transport emissions in a reasonable way. By applying the same calculation method for Beirut and its suburbs, the annual emissions obtained using the summer ratio are estimated to be 29 Gg higher than the annual emissions calculated based on winter ratio (23 Gg) (figure 12).

The measurements we conducted are more representative of Beirut and its suburbs (Greater Beirut Area). The emissions from the inventory regarding the road transport for Greater Beirut Area are also lower with a difference ranging from 20% for winter based emissions to 37% for summer based emissions.

The discrepancies for Lebanon as well as for Beirut and its suburbs using winter and summer ratios are comparable since the road transport is the dominating sector in the whole country and especially in the capital.

7.2. Comparison to global emission inventories (for the road transport sector)

The objective here is to assess the PMF estimated emissions to the global emission inventories for the road transport sector for 59 NMHCs and, where possible, for a common combustion tracer “ethylene” which is also an ozone precursor due to its reactivity towards the OH radical (Atkinson and Arey, 2003).

We estimated the annual total NMVOCs emissions for Lebanon by using a mean flux of total NMVOCs of 8 grids covering Lebanon surface area (ECCAD database) from the transportation sector from the global historical emission inventory ACCMIP (Atmospheric Chemistry and Climate Model Intercomparison Project) (Lamarque et al., 2010) for 2000 at a spatial resolution of 0.5°. The total annual NMVOCs transportation emissions from the ACCMIP are estimated to be 8 Gg, which is a factor of 6 to 10 lower than the annual emissions obtained via the PMF results (62 to 78 Gg) and the road transport national inventory emissions (48 Gg). This gap is expected to be a lower limit regarding the fact that PMF only consider 59 NMHCs. Nevertheless, when we apply the same method to another global inventory EDGARv4.2 (Emissions Database for Global Atmospheric Research) at a spatial resolution of 0.5°, the annual transportation emissions are estimated to be 23 Gg, which is a factor of 2 to 3 lower than the road transport national inventory emissions and the annual emissions obtained via the PMF results. This comparison also reveals that global inventories are not consistent between each other. Additionally, the global inventories do not describe the methodologies applied neither the data used (emission factors, NMVOC species considered…).

For a more detailed evaluation of speciation accuracy, we compared the emissions of an individual NMHC, ethylene, obtained by the PMF results according to equation (5), where “NMHC” represents ethylene, to the emissions from a speciated inventory having a monthly temporal resolution, over the same period, called MACCity. The MACCity global emission inventory has been developed within two European Commission funded projects (MACC and CityZen) as an extension of the ACCMIP and RCP8.5 (Representative Concentrations Pathways) emission inventories (Granier et al., 2011) at a spatial resolution of 0.5°. Based on the PMF results, the emissions of ethylene during July 2011 are estimated to 0.30 Gg and 0.25 Gg during January 2012 for Lebanon. The MACCity global emission inventory displays very low ethylene emissions from the transportation sector (ECCAD database) during July

2011 (0.05 Gg) and during January 2012 (0.04 Gg) for Lebanon (figure 12). The difference with the PMF results is by a factor of 6.

8. Conclusion

Measurements of 67 gaseous organic carbon have been performed at a sub-urban site in Beirut during two intensive field campaigns in summer 2011 and in winter 2012 within the framework of the ECOCEM project, resulting in a large and unique database, in order to improve our understanding of their major emission sources in the area.

The USEPA PMF v3.0 was applied to the winter and summer datasets in order to identify and quantify the source contribution. Six factors were then extracted in winter and five factors in summer. Source profiles established close to the main potential emitters by near field measurements (Salameh et al., 2014) were used to identify the PMF factors.

In winter, the six NMHC emission factors consist of traffic related sources including combustion mainly related to regional traffic, combustion related to local traffic and gasoline evaporation from vehicles accounting for 51%. In summer, the combustion and gasoline evaporation related to regional traffic contributed to 74% of which 57% are nocturnal. Additionally, an episodic gasoline evaporation source originating principally from the fuel storage facilities was identified in winter and in summer contributing to 21% and 16% respectively. The gas leakage mainly LPG contributes to 10% and the urban background to 18% of the ambient NMHC levels during winter. A factor characterizing local background was identified in summer contributing to 10%. Having similar species composition profiles, we found a difficulty in discerning the combustion related to power generators, running on diesel oil, within the combustion sources, hence, additional species (e.g. PM2.5 species measured with high temporal resolution) may have helped to extract this source.

Overall, the gasoline evaporation contribution (related to traffic and the episodic point source) was found to be a large contributor to the NMHC ambient concentrations: 43% in winter and 46% in summer.

Control measures should be developed to mitigate the NMHC emissions resulting from traffic-related sources (combustion and gasoline evaporation).

Verification of the reported emission inventory for Lebanon and Greater Beirut Area can assist in the evaluation of future control strategy effectiveness. The comparison of the modeled results to the emission inventory shows an agreement regarding the load of the on-road mobile source, by considering the vehicle exhaust and gasoline evaporation linked to traffic, in both seasons. Nevertheless, the gasoline evaporation related to traffic is lower in the inventory (17%) by considering PMF summer results (30%) suggesting an underestimation of the inventory. We assessed also the accuracy of several global inventories for the transportation sector and high discrepancies up to a factor of 10 still remain between the emission inventories.

To maximize current resources, continuous measurement of NMHC species and the implementation of ground based monitoring networks for ambient air quality monitoring and assessment are essential in order to conduct source apportionment which would be of less uncertainty and of greater help in establishing effective NMHC, ozone and PM abatement strategies in Lebanon.

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Table 1: Mathematical diagnostic for the PMF results.

	Summer	Winter
n (samples)	298	179
m (species)	59	59
k (factors)	5	6
Q (model)	5670	1728
$NMHC_{modelled}$ vs $NMHC_{measured}$ (R)	0.97	0.99
F_{peak}	-0.5	0
Mean ratio (modelled vs. measured)	1.05	0.99
Number of species with $R^2>0.75$ for modeled vs measured	39	29

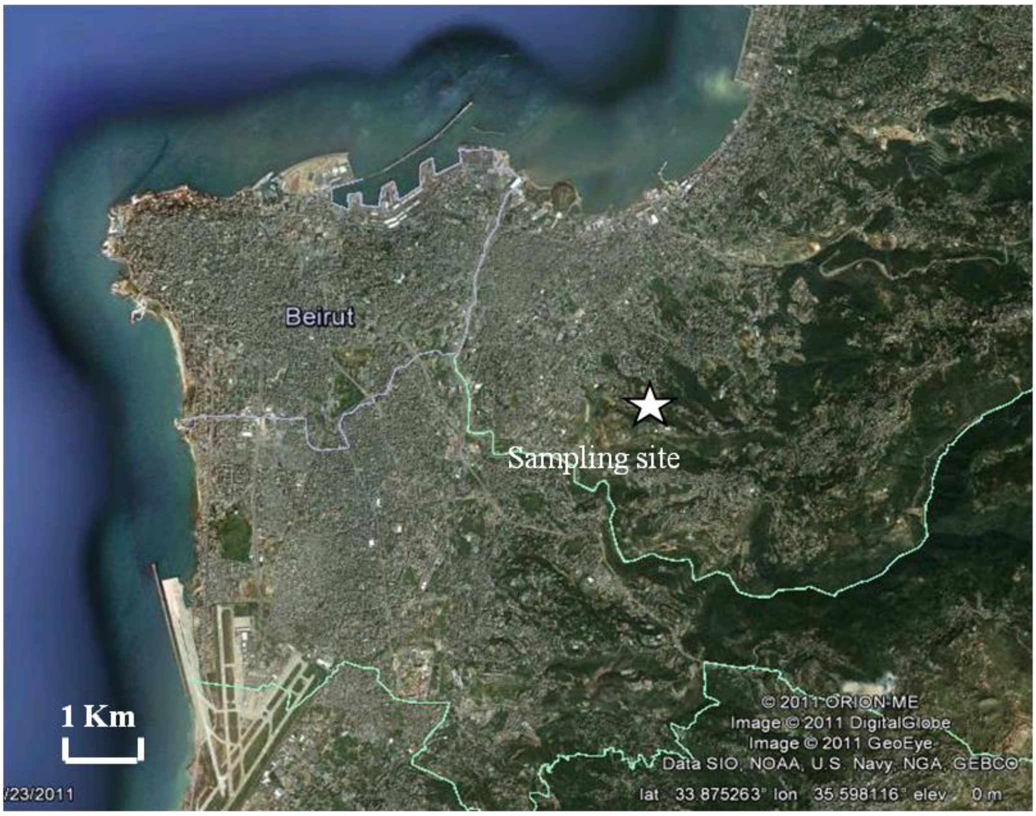

Figure 1: Sampling site in the eastern suburb of the city of Beirut.

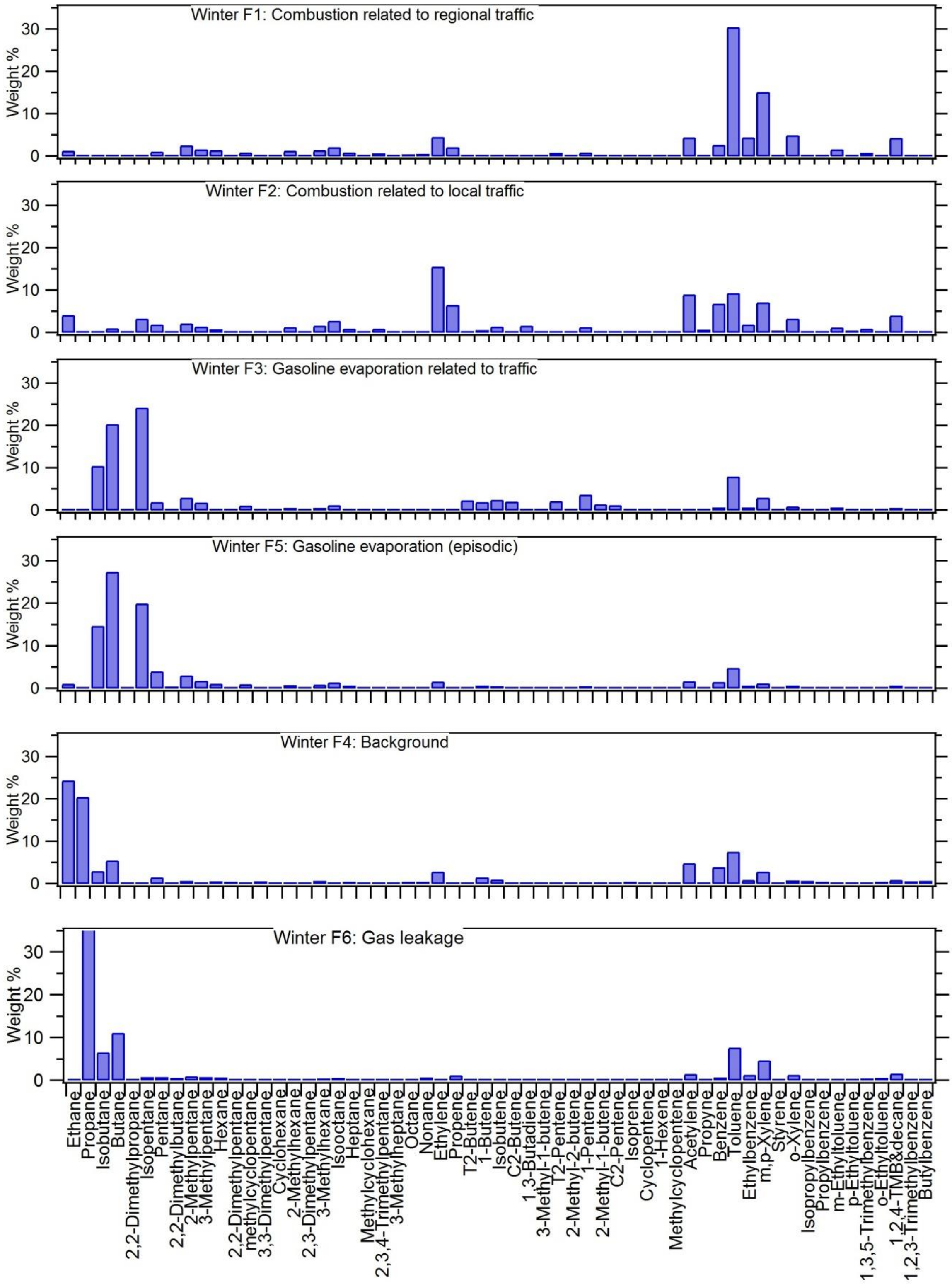

Figure 2: Source composition profiles (relative abundance of specific compounds within each factor in weight %) in winter 2012.

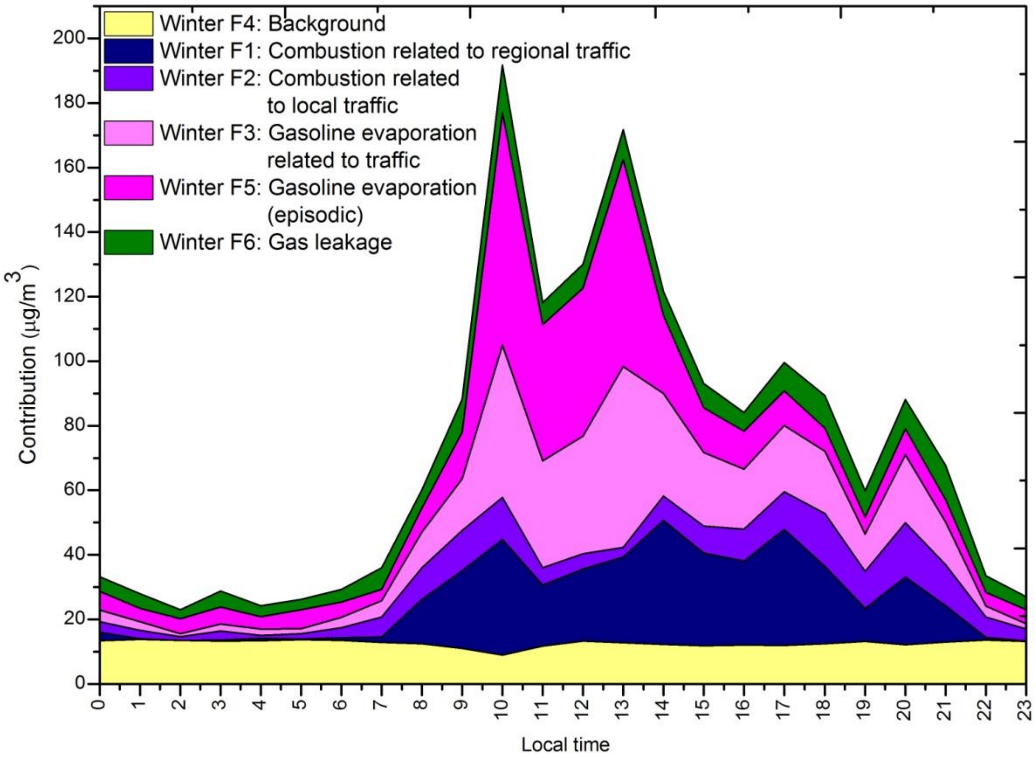

Figure 3: Diurnal variation of source contribution (µg/m^3) to NMHC concentrations in winter 2012.

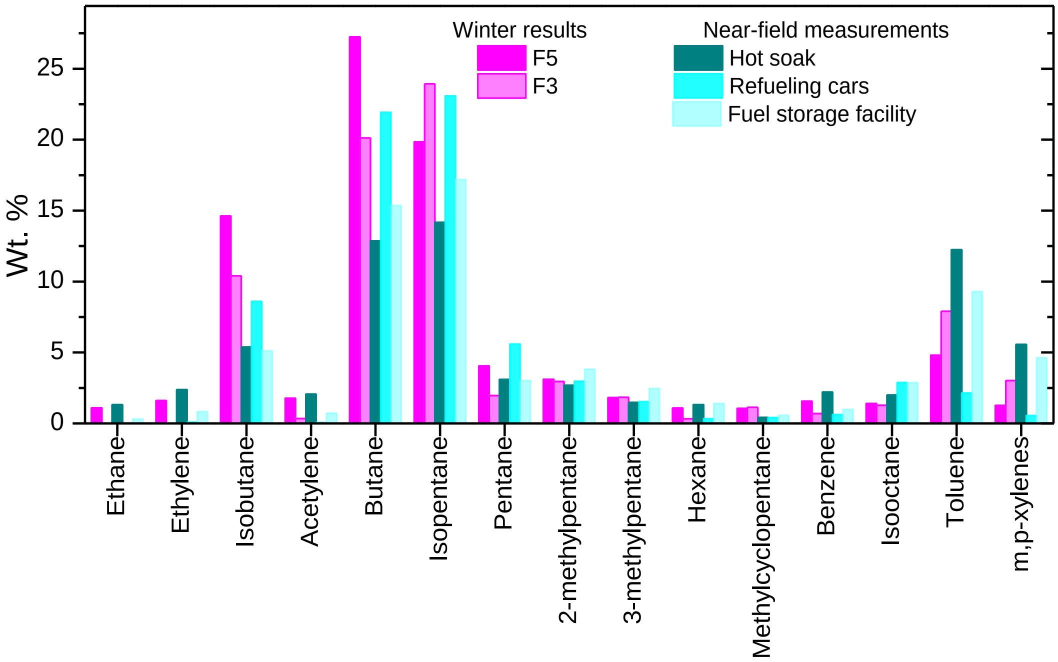

Figure 4: Composition profiles (weight %) of two gasoline evaporation factors (F5 and F3) in winter compared to typical gasoline evaporation profiles established by near field measurements (Salameh et al., 2014).

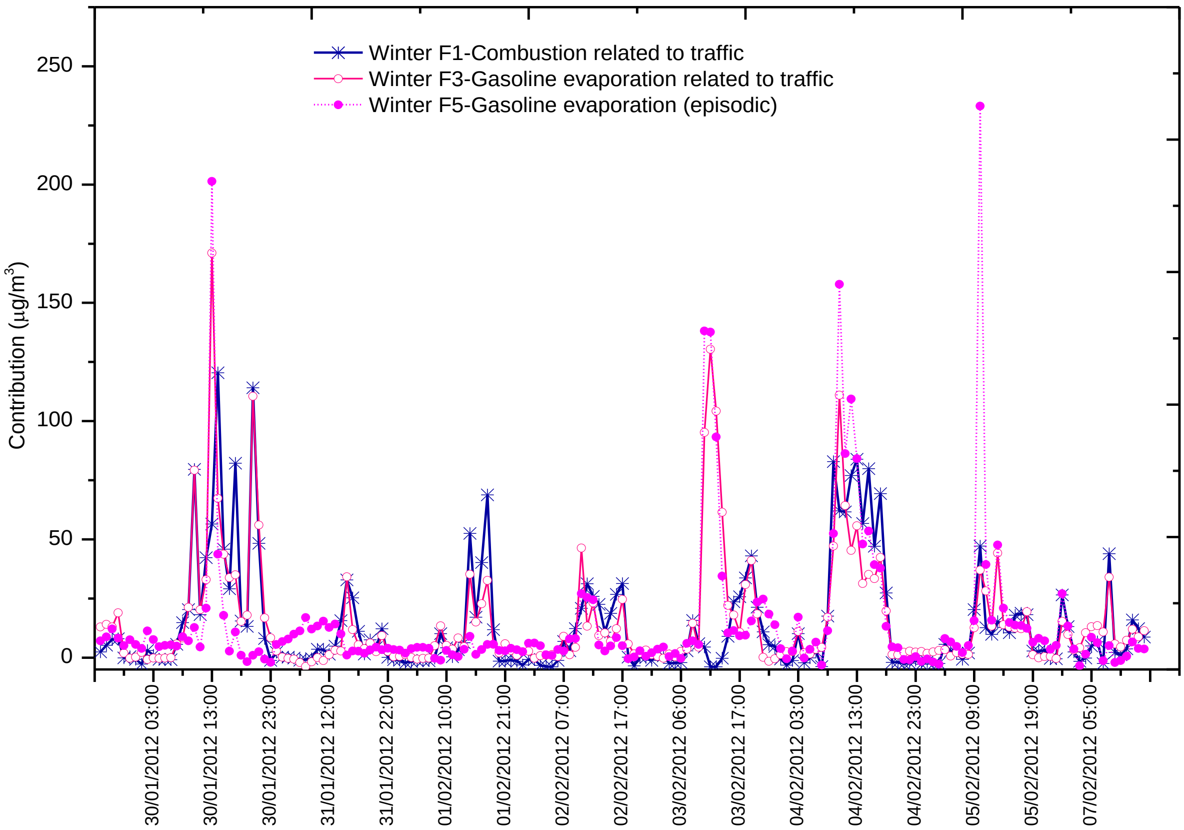

Figure 5: Time series of winter gasoline evaporation (F3 and F5) and combustion related to traffic factor contribution (F1) in µg/m^3.

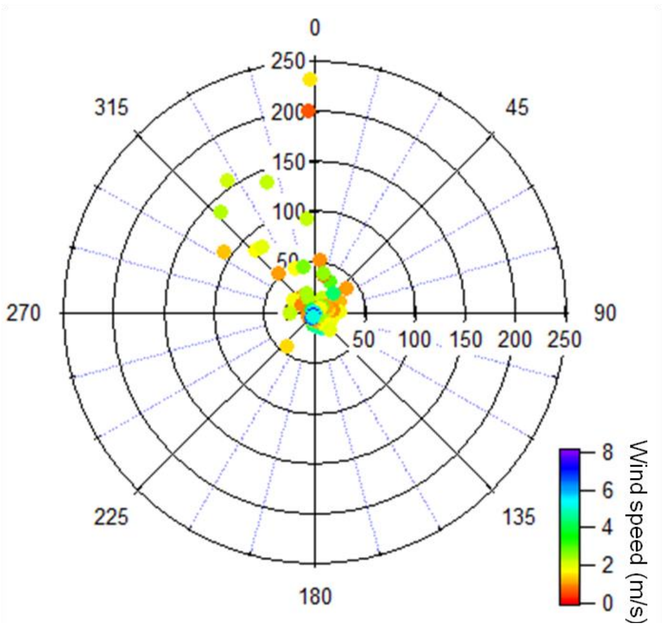

Figure 6: Pollution rose of factor 5 contribution (µg/m^3) in winter.

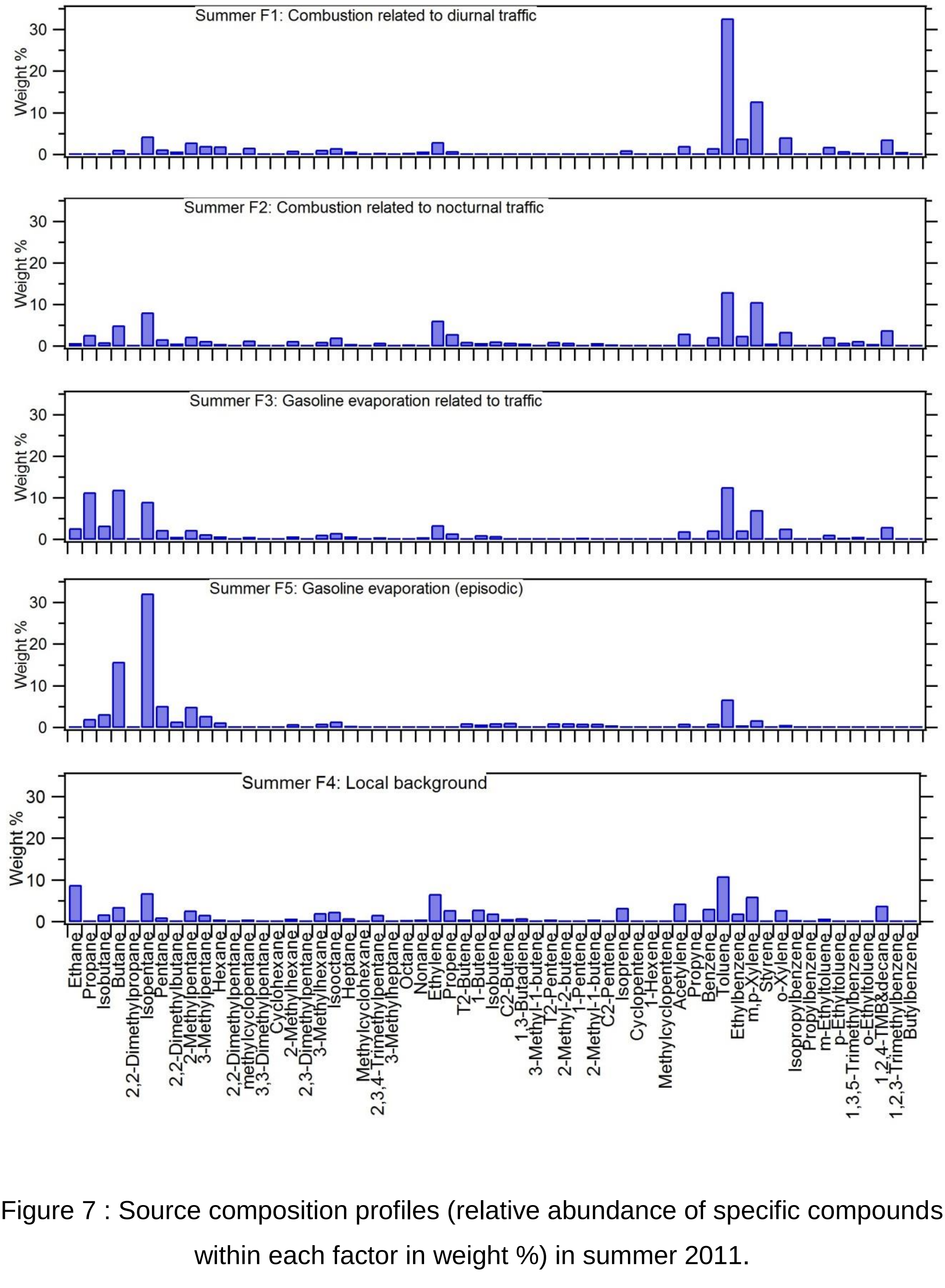

Figure 7 : Source composition profiles (relative abundance of specific compounds within each factor in weight %) in summer 2011.

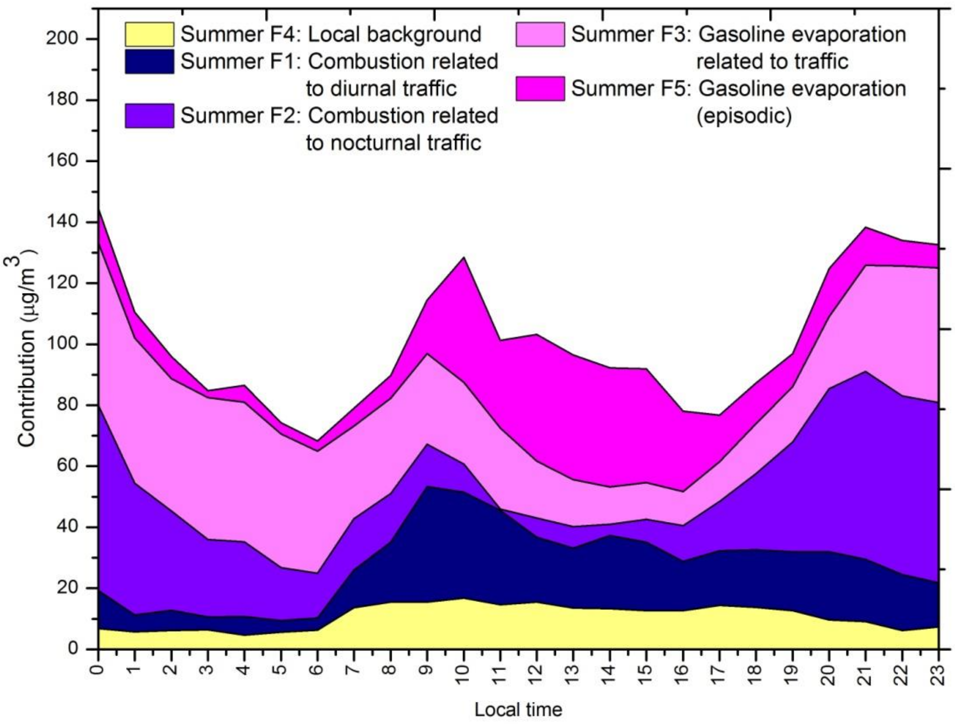

Figure 8 : Diurnal variation of sources contribution (µg/m^3) in summer 2011 (local time).

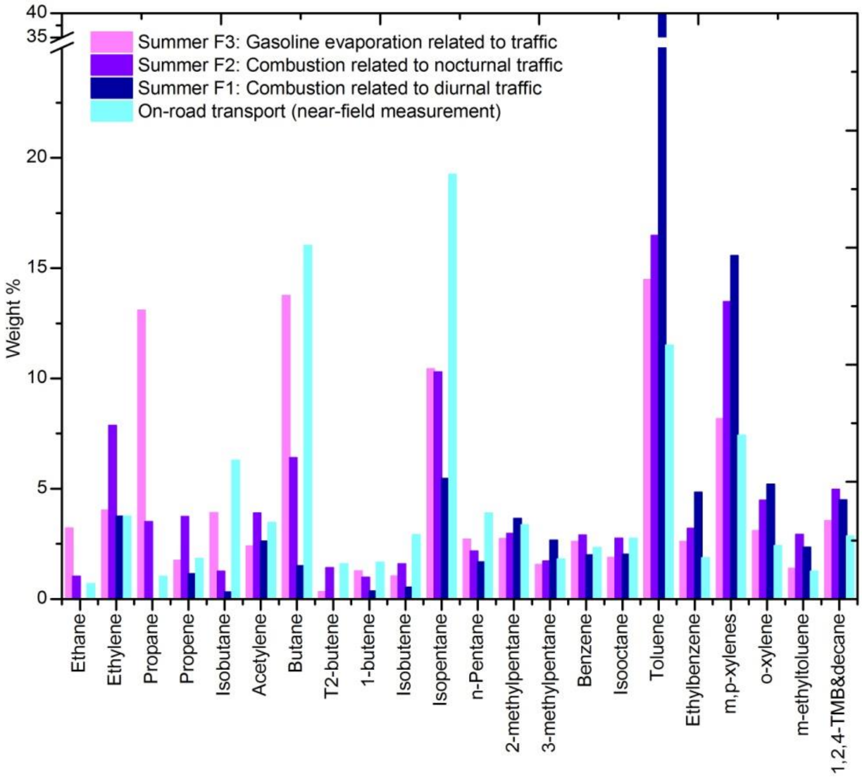

Figure 9: Source composition profiles (weight %) of gasoline evaporation (F3) and combustion related to diurnal and nocturnal traffic (F1 and F2) in summer compared to the on-road transport profile established by near field measurements (Salameh et al., 2014).

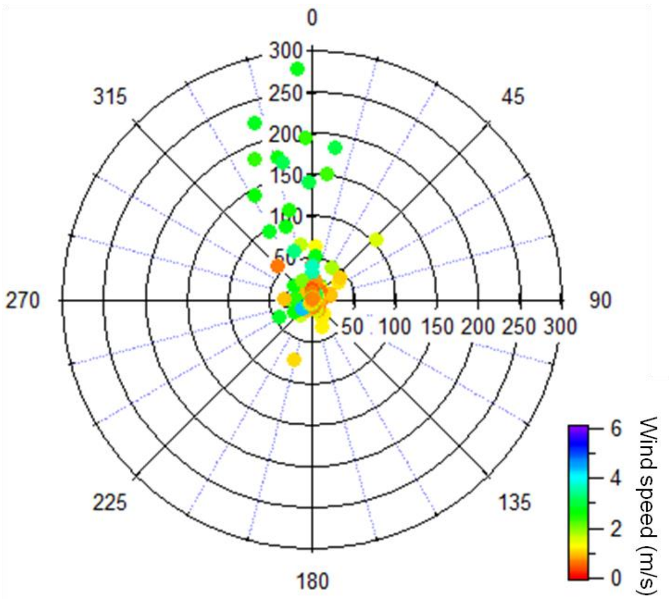

Figure 10: Pollution rose of factor 5 contribution (µg/m^3) in summer.

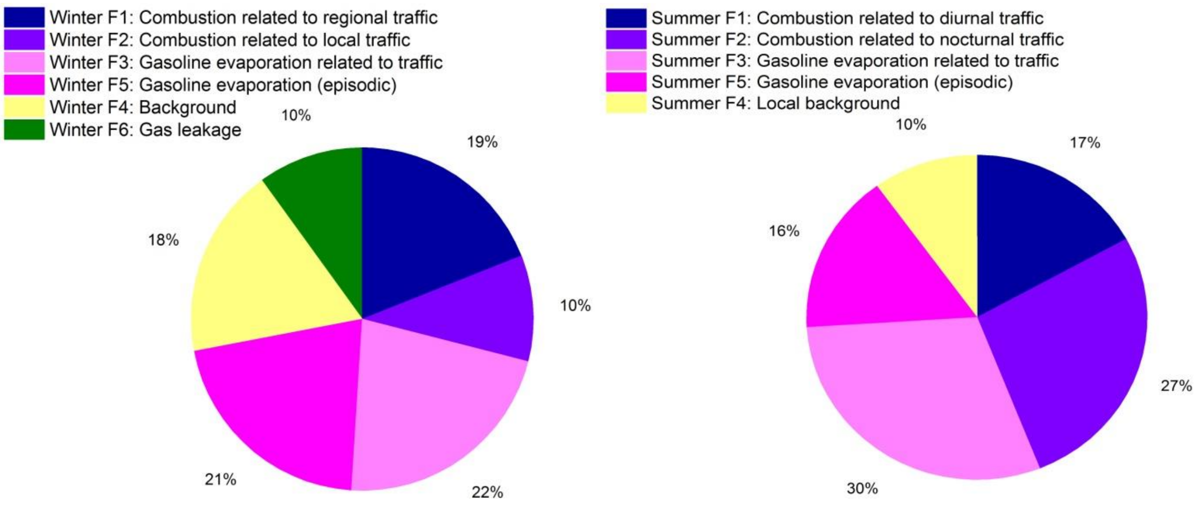

Figure 11: Sources contribution (weight %) in winter and in summer.

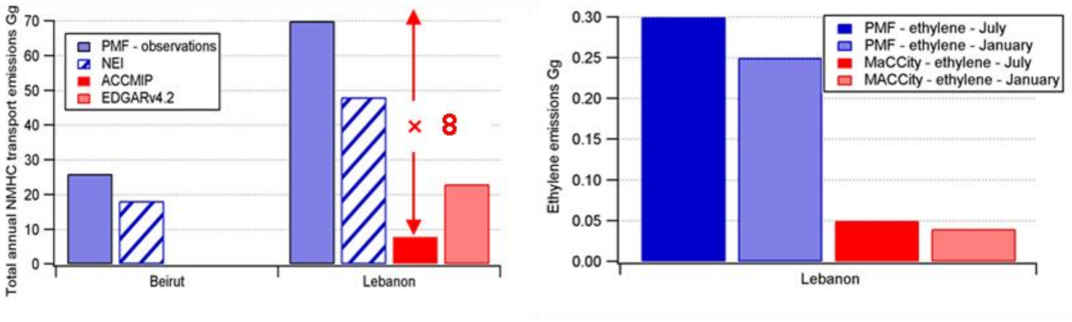

Figure 12: Comparison of the PMF results and emission inventories (national and global) for the transport sector emissions.