Atmospheric Pollution Research 7 (2016) 971–979

Contents lists available at ScienceDirect

Atmospheric Pollution Research

journal homepage: http://www.journals.elsevier.com/locate/apr

Review article

Influence of boundary conditions and anthropogenic emission inventories on simulated O_3 and $PM_{2.5}$ concentrations over Lebanon

C. Abdallah [a, b, *], K. Sartelet [a], C. Afif [b]

[a] Centre d'Enseignement et de Recherche en Environnement Atmosphérique (CEREA), Ecole des Pont et Chaussées, Champs sur Marne, France
[b] 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

ARTICLE INFO

Article history:
Received 15 December 2015
Received in revised form
2 May 2016
Accepted 3 June 2016
Available online 17 June 2016

Keywords:
Air quality modeling
Particulate matter
Ozone
Middle East
Lebanon

ABSTRACT

This study investigates the influence of boundary conditions and anthropogenic emission inventories on the simulated O_3 and $PM_{2.5}$ concentrations over a middle-eastern country – Lebanon. The Polyphemus chemical transport model (CTM) is used over Lebanon to simulate O_3 and $PM_{2.5}$ concentrations. Comparisons to measurements at a sub-urban site of Beirut between 2 and 13 July 2011 show that O_3 is largely over-estimated when concentrations from a large-scale model are used as boundary conditions, as used in Waked et al. (2013). A global anthropogenic emission inventory (EDGAR-HTAP) is used with Polyphemus, in order to provide anthropogenic emissions for the Middle-East domain. Over Lebanon, sensitivity to emissions and to boundary conditions have been investigated. The comparison of EDGAR-HTAP to Waked et al. (2012) over Lebanon highlights high discrepancies between the inventories both in terms of emission estimates and spatial distribution. However, when studying the sensitivity to boundary conditions, O_3 is well modeled when a Middle-East domain and the Lebanon domain are nested and thus achieves better statistics. The observed concentration is 48.8 μg m^{-3} and the respective concentrations for the simulation using MOZART4 and the one using the Polyphemus/Middle-East are 154.8 and 65.1 μg m^{-3}. As for $PM_{2.5}$ which is less sensitive to regional transport than O_3, the influence of the boundary conditions on the $PM_{2.5}$ concentrations at the site of comparison is low. The observed concentration is 20.7 μg m^{-3}, while the modeled concentrations are 20.7 and 20.1 μg m^{-3} respectively.

Contents

* Corresponding author. Centre d'Enseignement et de Recherche en Environnement Atmosphérique (CEREA), Ecole des Pont et Chaussées, Champs sur Marne, France.
E-mail addresses: Charbel.abdallah@enpc.fr, Charbel.abdallah@net.usj.edu.lb (C. Abdallah).
Peer review under responsibility of Turkish National Committee for Air Pollution Research and Control.

http://dx.doi.org/10.1016/j.apr.2016.06.001

1. Introduction

Lebanon, a developing country of the Middle East, is facing multiple challenges related to ambient air quality problems resulting from climatic conditions, high anthropogenic emissions, and persistent air pollution conditions (Afif et al., 2008, 2009; Waked et al., 2013). However, the only legislation made for ambient air quality standards dates back to 1996 (MoE, 1996), and has not been updated since then. Nowadays, this legislation is considered as outdated in comparison to the current ambient air quality standards of the European Union (EU) and the World Health Organization (WHO), as it does not define standards for $PM_{2.5}$ concentrations, and tolerates higher concentrations than in the WHO guidelines (WHO, 2006) for yearly NO_2, PM_{10}, etc. Table 1 compares the air quality standards of the Lebanese legislation to those of the European legislation and those of the WHO guidelines. Until 2013, air quality in Lebanon was assessed through initiatives conducted by universities as part of their short-term research studies and focused mainly on Beirut (Afif et al., 2008, 2009; Waked et al., 2013, 2014). In 2013, the Ministry of Environment with the support of the United Nations Environment Programme (UNEP) and the United Nations Development Programme (UNDP) established the first phase of the national air quality monitoring network comprising five air quality monitoring stations. Data is not public yet. The achievement of the expansion of the air quality network is planned to increase the number of stations to sixteen in 2016.

In an effort to better understand and estimate pollutant concentrations over Beirut, a summer campaign with pollutant measurements (CO, NO_x, O_3, and $PM_{2.5}$) was performed in 2011. Simultaneously, Waked et al. (2012) put together the first emission inventory for both Lebanon (with a grid resolution of 5 km) and Beirut and its suburbs (with a grid resolution of 1 km). They used it in an air-quality modeling study using the Polyphemus chemical transport model (CTM) (Mallet et al., 2007) over two nested domains (Lebanon and Beirut) (Waked et al., 2013). Over Lebanon, their simulations showed a good spatial representation of the country's hotspots such as Beirut, Chekka, and Zahle. Over Beirut, the results were satisfactory for most of the pollutant concentrations measured during the summer campaign of 2011. However, simulations showed an over-estimation of ozone concentrations over Beirut when compared to measurements. This issue was resolved by dividing the boundary conditions for ozone of their coarsest domain (Lebanon) by a factor two. These boundary conditions were obtained from a global-scale model MOZART-4 (Model for OZone And Related chemical Tracers version 4) (Emmons et al., 2010). In order to simulate accurately pollutant concentrations over Lebanon and Beirut, boundary conditions of the simulations over Lebanon need to be improved. To that end, in this paper, simulations over a larger domain than Lebanon are performed using Polyphemus and are used as boundary conditions of the Lebanon simulation. Since an emission inventory over such larger domain is not available, a global emission dataset is used: EDGAR-HTAP (Janssens-Maenhout et al., 2012). This dataset has a grid resolution of $0.1^\circ \times 0.1^\circ$ and it uses modeling to estimate emissions over regions where emissions inventories are not available. In this global dataset, modeling is also used over certain regions such as Europe to improve the spatial resolution of available gridded emission inventories. Over Lebanon in EDGAR-HTAP, modeling is used, although a gridded emission inventory exists (Waked et al., 2012).

The objective of this paper is to improve air quality simulations over Lebanon. To that end, we study the sensitivity of the concentrations simulated over Lebanon to the anthropogenic emission datasets and to the boundary conditions used. The paper is organized as follows: in Section 2, the air-quality modeling system Polyphemus used in the simulations of this paper is briefly described. Section 3 presents the global emission dataset EDGAR-HTAP and the local emission dataset used over Lebanon. The impact of emission modeling in the global emission inventory EDGAR-HTAP is studied by comparison to local inventories over Europe and Lebanon, and the impact of the emission dataset used on pollutant concentrations simulated with Polyphemus is evaluated. The potential shortcomings of the EDGAR-HTAP emission dataset over Lebanon are estimated. Finally, Section 4 presents a Middle East simulation performed with Polyphemus and the impact of using it as boundary conditions of the Lebanon simulations.

Table 1
The current Lebanese Air Quality Standards (in $\mu g\ m^{-3}$) as found in the legislation of 1996 (MoE, 1996) along with the current standards from the European Union legislation (EU, 2008) and the World Health Organization guidelines (WHO, 2006).

Pollutant	Duration of exposure	Lebanon	EU	WHO
SO_2	10 min	–	–	500
	1 h	350	350	–
	24 h	120	125	20
	1 year	80	–	–
NO_2	1 h	200	200	200
	24 h	150	–	–
	1 year	100	40	40
O_3	1 h	150	–	–
	8 h	100	120	100
CO	1 h	30,000	–	30,000
	8 h	10,000	10,000	10,000
Lead	1 year	1	0.5	0.5
Benzene	1 year	16	5	excess lifetime risk of leukemia
TSP	24 h	120	–	–
PM_{10}	24 h	80	50	50
	1 year	–	40	20
$PM_{2.5}$	24 h	–	–	25
	1 year	–	25	10

2. Modeling set-up

The Polyphemus air quality modeling platform is used in this study with the CTM Polair3d (Mallet et al., 2007). The set-up is similar to Waked et al. (2013) for the Lebanon domain. A coarser domain is added for the Middle East region and comprises the Lebanon domain. This Middle East domain is added in order to study the influence of the boundary conditions of the Lebanon domain on the concentrations. In the default set-up used in this paper, the simulation over Lebanon uses concentrations from the simulation over the Middle East as boundary conditions. The simulation domains are presented in Fig. 1. The Middle East domain covers the east Mediterranean basin and the Arab peninsula. It extends from 21°E to 46°E and from 24°N to 41.5°N with a horizontal resolution of $0.25^\circ \times 0.25^\circ$. The Lebanon domain extends

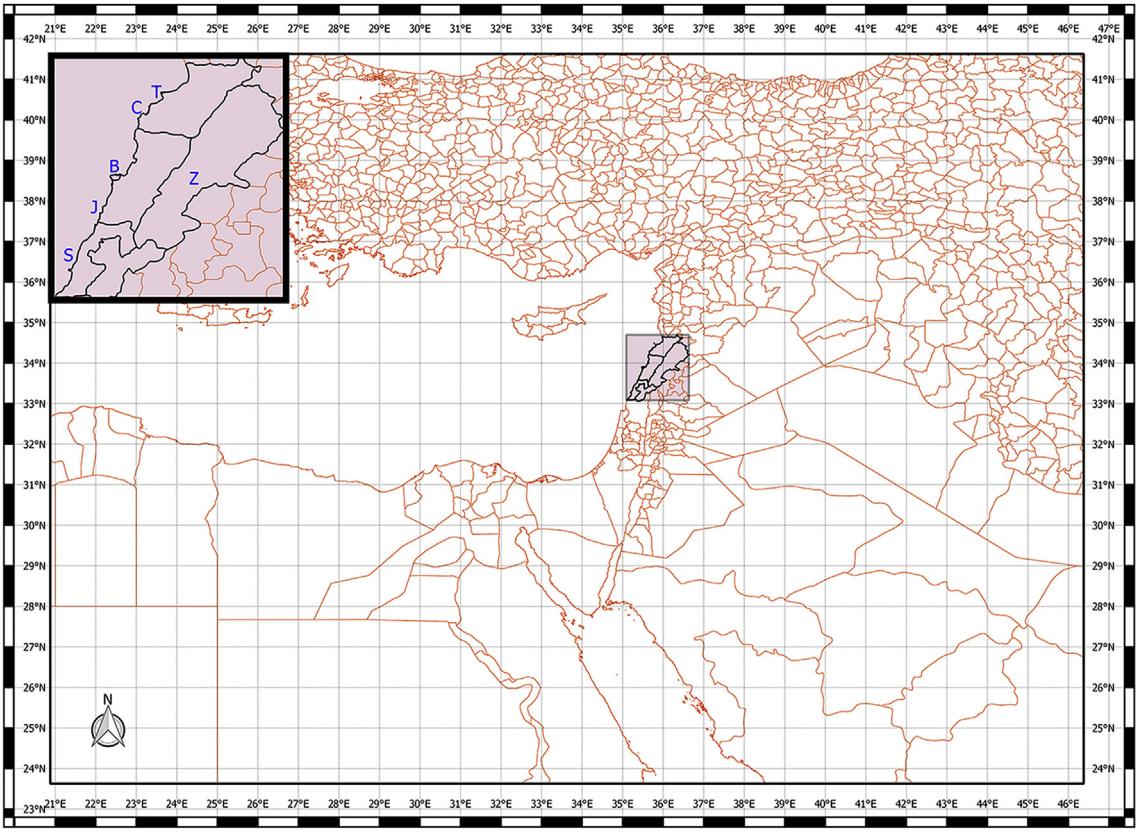

Fig. 1. The modeling domains used in this study. (B = Beirut, C = Chekka, J = Jiyeh, S = Tyre, T = Tripoli, Z = Zahle).

from 35.1175°E to 36.6575°E and from 33.103°N to 34.71°N with a horizontal resolution of 0.055° × 0.055°. Both domains have 9 vertical levels from the ground to 12 km height. Meteorological conditions are provided by ECMWF for the Middle East domain, and by WRF-ARW for the nested domain (Skamarock et al., 2008). WRF is driven by the input data from the National Centers for Environmental Prediction (NCEP) reanalysis meteorology datasets. The meteorological simulation uses the University of Washington Moist Turbulence scheme (Bretherton and Park, 2009) for Planetary Boundary Layer (PBL). The United States Geological Survey map is used over the Middle East/Lebanon domains. Biogenic emissions are calculated using Model of Emissions of Gases and Aerosols from Nature (MEGAN) (Guenther et al., 2006), which is designed for global and regional emission modeling. For anthropogenic emissions, EDGAR-HTAP version 1 is used over the Middle East domain, and either EDGAR-HTAP version 1 or Waked et al. (2012) inventories are used over Lebanon, as described in section 3. Initial and boundary conditions for the Middle East domain are extracted from the MOZART-4/GEOS-5 outputs that are gridded at a 1.9° × 2.5° horizontal resolution with 56 vertical levels. As specified on the MOZART-4/GEOS-5 website (NCAR, 2013), the simulation MOZART-4/GEOS-5 is driven by meteorological fields from the NASA GMAO GEOS-5 model and uses anthropogenic emissions based on David Streets' inventory for ARCTAS (Streets et al., 2003) and fire emissions from Fire Inventory from NCAR (FINN-v1) (Wiedinmyer et al., 2011).

The speciation of emissions is the same as in Waked et al. (2012) and Sartelet et al. (2007), except for the ratio of primary NO_2 over NO_x for traffic exhaust emissions. In Waked et al. (2012), similarly to Sartelet et al. (2007) over Europe, NO_x emissions were split into 90% of NO (in mass), 9.2% of NO_2 and 0.8% of HONO. Here, the NO_2/NO_x ratio for traffic exhaust is calculated for Lebanon based on the estimated values reported by Kousoulidou et al. (2008) for various emission control technologies by vehicle class and fuel type, and on the data from the vehicle count study performed in Beirut (Waked and Afif, 2012) and available data on the annual traveled distance per vehicle class (MOE/UNDP/GEF, 2015). The general method consists in estimating an average ratio for each category of vehicles weighted by the proportion of each control technologies first, and then in weighting these ratios by the total distance travelled for each category, as shown in Table 2. For Lebanon, an average NO_2/NO_x ratio of 4.5 is found, and NO_x emissions are split into 94.7% NO, 4.5% NO_2 and 0.8% HONO for traffic emissions.

Table 2
Lebanon fleet information and class specific NO_2/NO_x ratios considered in the calculations. PC stands for Passenger Cars, LDV-G for Low-Duty Vehicles using Gasoline, LDV-D for Low-Duty Vehicles using Diesel and HDV for Heavy-Duty Vehicles.

	PC	LDV-G	LDV-D	HDV
NO_2/NO_x	3.65	3.65	21	12.05
Number of vehicles	1,232,229	92,966	12,596	33,032
Average annual travelled distance per vehicle (Km)	12,000	25,000	25,000	50,000
Total annual travelled distance per vehicle class (Km)	14,786,748,000	2,324,150,000	314900000	1,651,600,000

Table 3 summarizes the inputs for the simulations. The model is the same as in Waked et al. (2013) and it is presented briefly hereafter. For gas-phase chemistry, a Carbon-Bond mechanism is used (CB05), it lumps organic species based on their types of carbon bonds (Yarwood et al., 2005). For aerosol modeling, the Size Resolved Aerosol Model (SIREAM) (Debry et al., 2007) is used with the Hydrophilic/Hydrophobic Organic model (H^2O) (Couvidat et al., 2012) for secondary organic aerosols (SOA) formation and ISORROPIA for inorganic aerosols thermodynamics (Nenes et al., 1998). SIREAM models coagulation and condensation/evaporation. In this paper, the simulations over the Lebanon domain are performed from 20 June 2011 to 20 July 2011, and a five-day spin-up time is added for all domains.

3. Sensitivity to the emission inventory

This section presents the global emission dataset that is chosen to perform air-quality simulations over the Middle East, and it compares it to local emission inventories. First, the local emission inventory of Waked et al. (2012) over Lebanon is briefly presented. Then, the choice of the global emission dataset is explained. Over Lebanon and the Middle East, the global emission dataset uses modeling to estimate emissions. However, over Western Europe, where local emission inventories are available and yearly updated, the global emission dataset uses modeling only for the spatial allocation of national emissions. The global emission dataset is compared to local emission inventories over Europe and Lebanon. The first comparison is performed over Western Europe, to estimate the impact of using modeling for the spatial allocation of national emissions. The second comparison is performed over Lebanon, where a local emission inventory is available but not used in the global dataset. The impact of the choice of the emission dataset on simulated pollutant concentrations is also evaluated.

3.1. The local emission inventory

In 2012, Waked et al. put together the first spatially resolved and temporally allocated anthropogenic emission inventory for Lebanon and its capital city Beirut for the year 2010 (Waked et al., 2012). The spatial resolution is 5 km × 5 km in the horizontal over Lebanon and 1 km × 1 km over Beirut and its suburbs. The spatial allocation is done using proxy subsets that consist of population density maps, point-sources locations, land cover, road networks and traffic density. The studied sectors include energy production, industrial activities, residential and commercial activities, solvent use, transportation, waste disposal and agriculture. The inventory includes the emissions of CO, NO_x, SO_2, VOC, NH_3, PM_{10}, and $PM_{2.5}$. The emission calculation methodology used differ between sectors, depending on the available data. Where local data is abundant, a bottom-up method was used as in the case of road transport (Waked and Afif, 2012), power plants and industrial plants. Similarly, where local data lacks, a top-down method was used.

Table 3
Summary of the input data used for each of the two domains studied.

Domain	Middle East	Lebanon
Initial conditions	MOZART-4/GEOS-5	Nested
Boundary conditions	MOZART-4/GEOS-5	Nested
Meteorology	ECMWF	WRF-ARW v3.5
Land use	USGS	USGS
Anthropogenic emissions	EDGAR-HTAP (2005)	Waked et al. (2012)
Biogenic emissions	MEGAN	MEGAN

3.2. The global emission inventory

Although many global emission datasets are publicly available (ACCMIP, ARCTAS, EDGAR4.2, EDGAR-HTAP, RCP), not all of them may be used to simulate air quality over the Middle East with Polyphemus. A global anthropogenic emission dataset is needed for the year 2011 or an adjacent time period, unlike ACCMIP which is compiled for the years 1850 till 2000 and used to estimate future scenarios. The data needs to be provided with a sectorial distribution, which is not the case of ARCTAS, in order to apply temporal profiles, which represent seasonal, monthly and daily source activity variations. Also, the data needs to include most commonly emitted pollutants (CO, NO_x, SO_x, NH_3, NMVOC, PM_{10}, and $PM_{2.5}$) related to air quality models. Furthermore, the spatial resolution of the data is important in deciding the inventory to be used. The global inventories EDGAR4.2 and EDGAR-HTAP are considered suitable for this study, as they satisfy the criteria listed above. Note that in the case of ARCTAS, which is used in the global model MOZART4, the inventory is based on the inventory EDGAR over the Middle East, but with a coarser spatial resolution than EDGAR-HTAP.

The Emissions Database for Global Atmospheric Research (EDGAR) is based on modeled emissions only, calculated through the use of surrogate global subsets such as population distribution, aviation paths, etc. EDGAR-HTAP emission inventory is a global inventory that contains emissions reported from countries/organizations such as the European Monitoring and Evaluation Program (EMEP, 2009) over Europe, combined with emissions calculated using EDGAR4.1 where reported emissions are not available. It should be noted that over Europe EDGAR-HTAP uses national totals from the EMEP emission inventory (and not the EMEP gridded values) that are then re-allocated spatially using EDGAR4.1 proxy subsets (Janssens-Maenhout et al., 2012). This results in small differences between both inventories. The first version of EDGAR-HTAP is available for the years 2000–2005. In this study, we use the emissions from 2005 for both Europe and the Middle East domains.

3.3. Emission dataset comparisons

3.3.1. Over Western Europe

Over Europe, EDGAR-HTAP uses national totals from the EMEP emission inventory, that are then re-allocated spatially using EDGAR4.1 proxy subsets to get a spatial resolution of 0.1° × 0.1°. This resolution is higher than the resolution of the EMEP gridded emission inventory (0.5° × 0.5°). The impact of the spatial allocation algorithm over Europe can be estimated by comparing the EDGAR-HTAP dataset to the EMEP gridded emission inventory, interpolated to a 0.1° × 0.1° spatial resolution. Fig. 2 shows the emissions of NH_3, NO_2 and $PM_{2.5}$ from EDGAR-HTAP and EMEP, as used in Polyphemus. In the case of NO_2, emissions from EDGAR-HTAP are lower over cities than emissions from EMEP, confirming that the spatial allocation is population dependent. In the case of NH_3, the spatial variations of EDGAR-HTAP emissions are higher than those of EMEP. EDGAR-HTAP emissions are higher in some areas such as western France, where for common years a comparison of the data showed that although both inventories have the same totals over the same zone, the cellular distribution differs. This is due to the fact that the spatial allocation of NH_3 depends on agricultural activities and its corresponding subsets such as the distribution of grass, cattle, rural population, etc. Also, it is noted that some gaps exist in EMEP emissions, especially for some Eastern-European countries, such as Azerbaijan, Romania, and Ukraine. Over these countries, the data is gap-filled with EDGARv4.1 emissions, which explains the differences near Hungary and

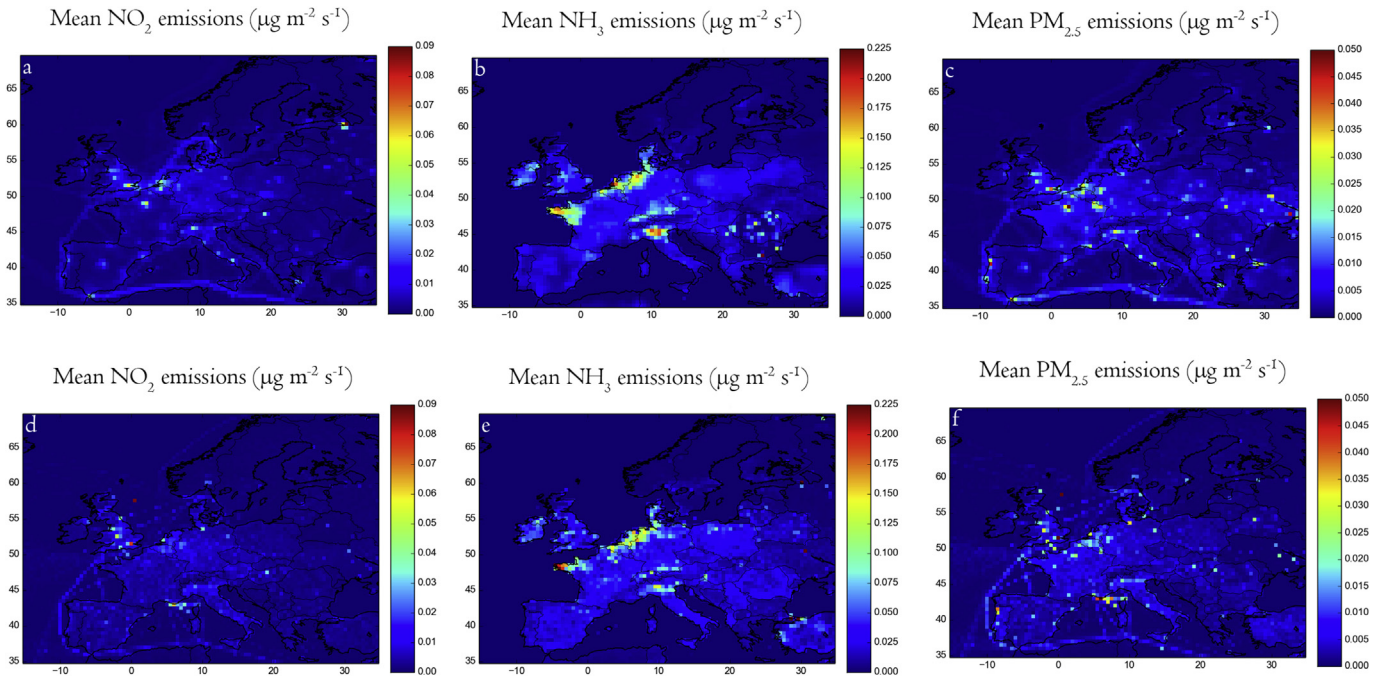

Fig. 2. Spatial distribution of mean emissions (μg m^{-2} s^{-1}) for the simulation period over Europe at a 0.1° × 0.1° horizontal resolution for EMEP NO_2 (a), NH_3 (b) and $PM_{2.5}$ (c); EDGAR-HTAP NO_2 (d), NH_3 (e) and $PM_{2.5}$ (f).

its surroundings. Similarly to NO_2, $PM_{2.5}$ emissions from EDGAR-HTAP differ from those of EMEP spatially. Both inventories highlight highly populated cities, yet EDGAR-HTAP resolution of 0.1° × 0.1° allows a better highlighting of secondary cities such as Valence, Marseilles, Seville, etc. Also, the sectorial analysis of EDGR-HTAP data shows some discrepancies between both inventories, especially over-seas where navigation routes differ between both inventories. In this case, NO_2 and $PM_{2.5}$ emissions highlight those differences and strong emissions are observed where navigation routes intersect such as the line connecting the island of Corsica to Marseille in southern France, or the high emitting cells in the North Sea near Scotland.

3.3.2. Over Lebanon

Over Lebanon, the emission inventory developed by Waked et al. (2012) has a horizontal resolution of 5 km × 5 km. Table 4 compares the totals for pollutant emissions from the EDGAR-HTAP inventory for the year 2005 and Waked et al. inventory for the year 2010. This comparison shows that EDGAR-HTAP overestimates the total emissions of NH_3 and SO_x emissions by a factor about 3, underestimates CO, PM_{10} and $PM_{2.5}$ by a factor between 2 and 3, while NO_x and NMVOC are well estimated. In addition to that, the calculated emissions have different spatial and temporal distributions. Fig. 3 shows emission maps of NO_x, NH_3 and $PM_{2.5}$ for Lebanon from both EDGAR-HTAP and Waked et al. inventory over Lebanon. In Waked et al. inventory, the maps show that the spatial distribution of NO_x emissions is more balanced over the domain, highlighting major road axes and highly emitting point sources. EDGAR-HTAP NO_x emissions from coastal roads along with the capital and its suburbs are spatially distributed similarly to Waked et al. inventory, although they are strongly under-estimated. Furthermore, in EDGAR-HTAP, the highly emitting industrial sources are not well represented. Industrial areas such as Chekka (see point C in Fig. 1), which is known for hosting major cement and fertilizer industries, in addition to a major highway and multiple quarries, or Jiyeh-Sibline area (see point J in Fig. 1), which also hosts a cement factory along with a public power plant, are strongly under-estimated in EDGAR-HTAP compared to Waked et al. inventory. In fact, the emissions of NO_x and $PM_{2.5}$ from Chekka and Jiyeh-Sibline account for most industrial emissions in Waked et al. inventory, but they are largely under-estimated in EDGAR-HTAP. A highly emitting industrial point emission in EDGAR-HTAP is located in the south eastern side of Beirut, which is not known as an industrial location in Waked et al. inventory. On top of uncertainties in the spatial allocation of industries and the under-estimation of road traffic, the relative part of industry and road traffic is represented differently in EDGAR-HTAP and in Waked et al. inventory. In fact, Waked et al. inventory shows that 52% of the annual NO_x emissions over Lebanon originate from on-road transport, while this percentage is only 28% in EDGAR-HTAP. For $PM_{2.5}$, the spatial distribution of the Waked et al. inventory highlights the high emissions along the coast-line, the capital and inland emissions. Similar to NO_x, $PM_{2.5}$ emitted from industrial sources is not well represented in EDGAR-HTAP, and traffic emissions are under-estimated. As detailed in Section 3.2, Waked and Afif (2012) applied the EMEP tier 3 method for traffic emissions in Beirut after conducting traffic counts, to characterize the volume and the composition of the traffic. In Beirut, they combined their manual

Table 4
Lebanon annual emissions from EDGAR-HTAP and Waked et al. inventories.

Pollutant inventory	CO (Gg)	NO_x (Gg)	SO_x (Gg)	NH_3 (Gg)	NMVOC (Gg)	PM_{10} (Gg)	$PM_{2.5}$ (Gg)
EDGAR-HTAP (2005)	192.5	68.8	184.5	14.1	65.6	5.0	4.4
Waked et al. (2012)	554.3	73.0	61.9	3.8	56.1	12.8	10.0

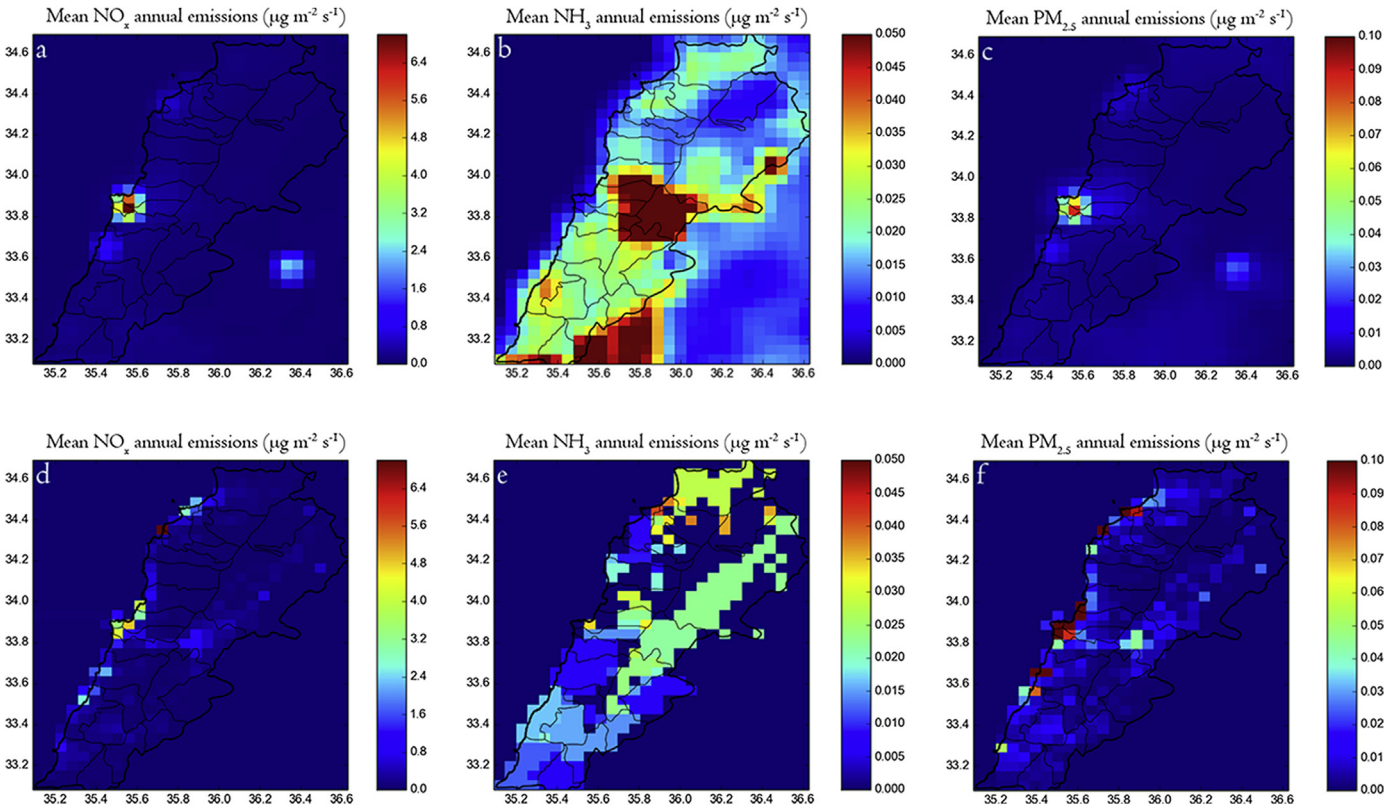

Fig. 3. Spatial distribution of annual mean emissions (μg m^{-2} s^{-1}) over Lebanon at a 0.055° × 0.046° horizontal resolution for EDGAR-HTAP NO_x (a), NH_3 (b) and $PM_{2.5}$ (c); Waked et al. NO_x (d), NH_3 (e) and $PM_{2.5}$ (f).

traffic count with different automatic counting studies. Over Lebanon, they scaled Beirut emissions using statistics based on origin-destination trips which are the number of daily trips that occur between each origin zone and each destination zone. EDGAR-HTAP estimated traffic emissions using an approach based on the total fuel consumed for transportation combined with estimation of fleet composition and emission factors. The data is spatially allocated afterwards based on the road network and the population density. Many sources of uncertainties originate from the global datasets used in the estimations of key factors such as the quantity of fuel, the distribution between diesel and gasoline vehicles and more generally the fleet composition. For population dependent sectors, such as non-industrial activities and waste disposal sectors, another important difference between the inventories is the base year for which they were made due to the continuous increase in the population. Since EDGAR-HTAP was made for the year 2005, the underestimation observed in these sectors can be related to the population estimates, compared to Waked et al. inventory, which was made for the year 2010. The Centre for International Earth Science Information Network population estimates (CIESIN et al., 2005) used in EDGAR-HTAP for the year 2005 showed a total population of 3.77 million people for Lebanon, while Waked et al. estimated the total population to 4.28 million people for the year 2010; which indicates a relative increase of 13.5% in total population, thus explaining some of the differences. For NH_3, EDGAR-HTAP has higher values than Waked et al. (2012), and a different spatial distribution. Fig. 3-d shows that inland NH_3 emissions in EDGAR-HTAP are concentrated around Zahle (see point Z in Fig. 1) and are much higher than other nearby regions of the Bekaa valley (3), which are also agricultural areas, as in the inventory of Waked et al. In other regions, such as South Lebanon, EDGAR-HTAP highlights important agricultural emissions from nearby countries that are included in the studied domain.

3.4. Impact of the emission dataset on concentrations

To assess the impact of the emission dataset on concentrations, and more particularly the impact of the emission modeling performed in the EDGAR-HTAP inventory, simulations are performed over both Europe and Lebanon. The simulations are compared to measurements and evaluated. The statistical indicators calculated are: the Mean Normalized Gross Bias (MNGB), the Mean Normalized Gross Error (MNGE), the Mean Fractional Error (MFE) and the Mean Fractional Bias (MFB). For O_3, the MNGE and MNGB are calculated with a cut-off of 80 μg m^{-3}. For hourly O_3, Russell and Dennis (2000) suggested that the performance criterion is met when both MNGB $\leq \pm 15\%$ and MNGE $\leq 35\%$. For PM, Boylan and Russell (2006) suggested that the performance criterion is met when both MFB $\leq \pm 30\%$ and MFE $\leq 50\%$, and that the model performance goal is met when both MFB $\leq \pm 60\%$ and MFE $\leq 75\%$.

3.4.1. Over Western Europe

The modeling set-up is the same as detailed in Section 2, except for the simulated domain and the emission inventories used.

The domain covers the western part of Europe (35°N − 70°N; 15°W − 35°E) with a resolution of 0.5° × 0.5°. 9 vertical levels are considered from the ground to 12 km (40 m, 120 m, 300 m, 800 m, 1500 m, 2400 m, 3500 m, 6000 m, and 12,000 m). The simulations are performed from 15 June 2009 to 31 July 2009. Two simulations are performed using the same input data except for anthropogenic emissions. The first simulation uses the EMEP gridded emission inventory, while the second uses the EDGAR-HTAP dataset.

To assess the performance of Polyphemus with the two emission inventories, the outputs from the simulations are compared to the observations of the Airbase network (Airbase, 2009). For hourly O_3, the average simulated concentration with EDGAR-HTAP is slightly lower than the simulated concentration with EMEP,

87.7 μg m^{-3} and 95.1 μg m^{-3} respectively, thus closer to the mean observed value of 63.3 μg m^{-3}. Both simulations verify the defined performance criterion. The simulation using EDGAR-HTAP has a MNGB and a MNGE of −3.2% and 14.8% respectively. The simulation using EMEP has a MNGB and a MNGE of 7.1% and 17.4% respectively.

For hourly $PM_{2.5}$, the simulation using EDGAR-HTAP leads to slightly higher concentrations than the simulation using EMEP, 12.0 μg m^{-3} and 11.7 μg m^{-3} respectively, thus closer to the mean observed value of 12.8 μg m^{-3}. Both simulations verify the defined performance criterion and goal, with a MFB and a MFE of 4.6% and 44.3% respectively for the simulation using EDGAR-HTAP, and a MFB of −1.4% and a MFE of 46.2% for the simulation using EMEP.

In summary, the simulation using EDGAR-HTAP leads to O_3 and $PM_{2.5}$ concentrations that are slightly closer to observations than EMEP. However, both the EDGAR-HTAP and the EMEP simulations verify the performance criteria for both O_3 and $PM_{2.5}$. Improvement of the spatial allocation of emissions in EDGAR-HTAP leads to an improvement in the statistics of comparisons to observations.

3.4.2. Over Lebanon

Using the set-up detailed in Section 2, two simulations are performed: one with the EDGAR-HTAP anthropogenic emission dataset, and the other one with the Waked et al. emission inventory for Lebanon. The simulations are performed for the period from 20 June 2011 to 20 July 2011, thus including the period where measurements are available (Waked et al., 2013).

The results from the simulations are compared to the available observations done during the summer of 2011, from July 2 to July 13, at a suburban site near Beirut: The Saint Joseph University - Faculty of Sciences campus (USJ site) in the region of Mansourieh (33.86 N, 35.56 E) distant by 6 km from the center of Beirut.

Table 5 summarizes the statistics of the comparison for both simulations. For hourly O_3, the defined performance criterion is verified for both the Waked et al. and the EDGAR-HTAP simulations. The Waked et al. simulation has a MNGB and a MNGE of 6% and 27.1% respectively. The EDGAR-HTAP simulation has a MNGB and a MNGE of 9% and 21% respectively. Both simulations over-estimate the measurements. The average modeled O_3 observed for the Waked et al. simulation is 65.1 μg m^{-3} and 84.3 μg m^{-3} in average for the EDGAR-HTAP simulation, against 48.8 μg m^{-3} for the measurements. For daily $PM_{2.5}$, both simulations also verify the defined performance criterion and goal. The Waked et al. simulation has a MFB of 0% and a MFE of 21%. The EDGAR-HTAP simulation has a MFB of −14% and a MFE of 28%. Both simulations tend to under-estimate the measurements. The simulated concentration using Waked et al. inventory is 20.1 μg m^{-3} at the observation site against 17.6 μg m^{-3} for the simulation using EDGAR-HTAP inventory, while the mean observed value is 20.7 μg m^{-3}.

Although the simulations with either of the emission inventory compare well to the measurements at the USJ site near Beirut, there are large differences outside Beirut, especially along the coastal line, as shown in Fig. 4. The spatial distribution of O_3 and $PM_{2.5}$ of Fig. 4 highlights the differences resulting from highly emitting industrial locations near Chekka, Jiyeh, Tripoli and Tyre, as well as the urban area of the capital – Beirut.

In summary, the results of this section show that the global emission inventory is well suited for domains, such as Europe, where regional emission inventories exist. In the case of modeling domains where no regional or national inventories exist, the estimation of national totals can be difficult, as well as the spatial allocation and sectors distribution. Accordingly, the evaluation of national totals without a good proxy dataset could be misleading. Looking at the statistics obtained with both emission inventories compared to the observations at a sub-urban location near Beirut, both inventories performed rather well despite the spatial allocation differences discussed earlier. However, large differences are observed on O_3 and $PM_{2.5}$ concentrations, especially along the coastal line and close to highly emitting industrial locations. Therefore, further work on emission inventories should focus on implanting national inventories that exist in global emission inventories, even though they are not reported to international organizations. For a better model performance assessment, further work should focus on acquiring measurements from more than one station in order to review the performance of the set-up over the whole country and different station types.

4. Sensitivity to boundary conditions

In this section, the simulations are performed over the two nested domains defined in Fig. 1 and detailed in Section 2. In the default simulation, the Middle East simulation is used as boundary conditions of the Lebanon domain. To assess the impact of boundary conditions over Lebanon, another set of simulations is performed using MOZART-4/GEOS-5 as boundary conditions for Lebanon domain, similarly to the simulations performed in Waked et al. (2013).

The results from the finest domain (Lebanon) are compared to the measurements performed at the USJ suburban site near Beirut during the summer of 2011, from July 2 to July 13. The simulation over Lebanon that uses boundary conditions from the Middle East simulation is referred to as Lebanon-Polyphemus, and the simulation over Lebanon that uses boundary conditions from MOZART-4/GEOS-5 is referred to as Lebanon-MOZART4. Table 6 summarizes the statistics of both simulations. For hourly O_3, the results of Lebanon-MOZART-4 are higher than those of Lebanon-Polyphemus with 154.8 μg m^{-3} and 65.1 μg m^{-3} respectively compared to an observed value of 48.8 μg m^{-3}. The performance criterion is not satisfied for the Lebanon-MOZART-4 simulation with a MNGB of 117% and a MNGE of 122.3%. However, the performance criterion is satisfied using Lebanon-Polyphemus: the MNGB and MNGE are 6% and 27.1% respectively. Unlike O_3, both simulations perform well for $PM_{2.5}$. The performance criterion is satisfied for the Lebanon-MOZART4 simulation, which estimates well the $PM_{2.5}$ concentrations, with a mean modeled concentration of 20.7 μg m^{-3} which is equal to the mean observed concentration of 20.7 μg m^{-3}; the MFB and the MFE are 2% and 20%. The Lebanon-Polyphemus also

Table 5
Statistical performance evaluation over Lebanon of hourly O_3 and daily $PM_{2.5}$ for the simulations using EDGAR-HTAP and Waked et al. inventories.

Emission inventory	O_3 hourly		$PM_{2.5}$ daily	
	EDGAR-HTAP	Waked et al.	EDGAR-HTAP	Waked et al.
Mean observed (μg m^{-3})	48.8	48.8	20.7	20.7
Mean modeled (μg m^{-3})	84.3	65.1	17.6	20.1
MNGB (%)	**9**	**6**		
MNGE (%)	**21**	**27.1**		
MFB (%)			**−14**	**0**
MFE (%)			**28**	**21**

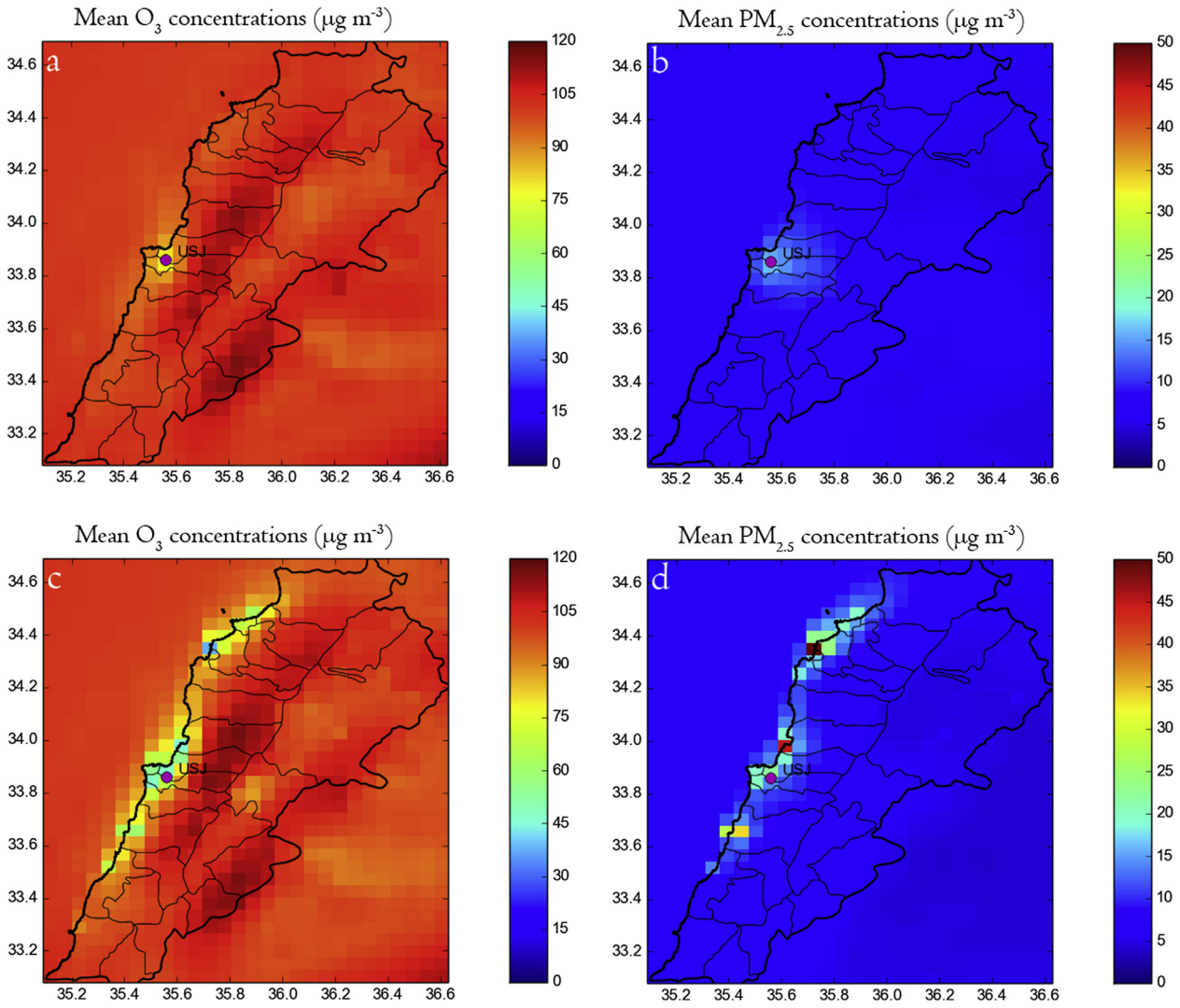

Fig. 4. Modeled average O_3 and $PM_{2.5}$ concentrations (μg m^{-3}) from using EDGAR-HTAP (a, b) and Waked et al. inventories (c, d) over Lebanon.

performs well: the mean modeled concentration is 20.1 μg m^{-3}; the MFB and the MFE are 0% and 21% respectively. The differences between O_3 and $PM_{2.5}$ concentrations emphasizes that at the USJ site near Beirut, O_3 is strongly impacted by regional transport, whereas the formation of $PM_{2.5}$ is mostly local.

The results of this section highlight the importance of using a simulation over the Middle East as boundary conditions of the Lebanon simulation, rather than the concentrations simulated by a large-scale model for simulating O_3 concentrations. The reason could relate to the set-up of the global-scale model MOZART-4 that has a 1.9° × 2.5° horizontal resolution, which is much coarser than our domain resolution over Lebanon (0.055° × 0.055°). Another reason for discrepancies could be related to the emission inventory used. This is however less likely, because both ARCTAS, which is used in MOZART4 and EDGAR-HTAP, which is used in Polyphemus, are based on EDGAR emission inventory over the Middle East.

5. Conclusion

To improve air quality simulations over Lebanon, sensitivity simulations to the anthropogenic emissions datasets and to the boundary conditions used were performed. The sensitivity simulations allowed us to identify the "good practice" to correctly model ozone and particulate matter concentrations over Lebanon.

Table 6
Statistical performance evaluation over Lebanon for hourly O_3 and daily $PM_{2.5}$ for the simulations using MOZART-4/GEOS-5 and Polyphemus boundary conditions.

Boundary conditions	O_3 hourly		$PM_{2.5}$ daily	
	MOZART-4	Polyphemus	MOZART-4	Polyphemus
Mean observed (μg m^{-3})	48.8	48.8	20.7	20.7
Mean modeled (μg m^{-3})	154.8	65.1	20.7	20.1
MNGB (%)	**117**	**6**		
MNGE (%)	**122.3**	**27.1**		
MFB (%)			**2**	**0**
MFE (%)			**20**	**21**

Comparisons to measurements were performed at one observation site in Beirut suburb (USJ site).

Two nested simulations were defined to model concentrations at the USJ site: one over the Middle East and one over Lebanon. Because O_3 is strongly impacted by regional transport, using boundary concentrations of the Lebanon domain from a large-scale model led to a strong over-estimation of O_3 concentrations at USJ. However, O_3 was well modeled when the Middle East simulation was used as boundary conditions.

Anthropogenic emissions from a global-scale emission inventory were used over the Middle East. Over Lebanon, either a local emission inventory or the global-scale emission inventory was used. For O_3 and $PM_{2.5}$, the performance criteria for comparison to measurements at the USJ site for the simulations over Lebanon were verified when the global-scale emission inventory was used as well as when the local emission inventory was used. However, using the global-scale emission inventory EDGAR-HTAP led to high spatial differences in the modeled O_3 and $PM_{2.5}$ concentrations, compared to the local emission inventory. These differences between the global-scale emission inventory EDGAR-HTAP and local emission inventories were investigated. Over Europe, improvement of the spatial allocation of emissions in EDGAR-HTAP compared to a nation-based emission inventory (EMEP) leads to an improvement in the statistics of comparisons to observations. However, over regions where nation-based emission inventories are not available (or not used), as over Lebanon, the global-scale emission inventories are usually based on modeling only, leading to high uncertainties in national totals.

Over Lebanon, the differences between EDGAR-HTAP and the local emission inventory, in terms of national totals and spatial allocation, highlighted high uncertainties both in terms of spatial allocation and emission quantification. National total emissions of EDGAR-HTAP are higher than those of the local emission inventory for NH_3 and SO_x by a factor about 3, and lower for CO, PM_{10} and $PM_{2.5}$ by a factor between 2 and 3, while they are similar for NO_x and NMVOC. The differences found in the industrial and transport sectors for NO_x and $PM_{2.5}$ emissions highlighted high uncertainties on the activity data as well as the choice of the emission factors used for power plants, cement factories, and on road emission sources. NH_3 differences highlighted furthermore the uncertainties in the spatial allocation of agricultural emissions in the global inventories.

Further work should focus on simulating more recent time periods, where common pollutants and meteorological fields are monitored continuously by the newly deployed national air quality monitoring network all over the country.

Acknowledgments

Funding for this study was provided by Ecole des Ponts ParisTech, the Lebanese National Council for Scientific Research and the Saint Joseph University (Faculty of Sciences).

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