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An emission processing system for air quality modelling in the Mexico City metropolitan area: Evaluation and comparison of the MOBILE6.2-Mexico and MOVES-Mexico traffic emissions

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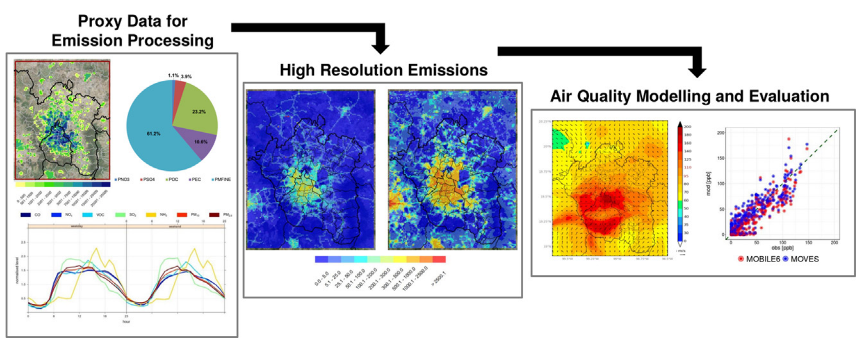
HIGHLIGHTS

- A tool to create air quality model-ready emissions for Mexico City is presented.
- The results are discussed and evaluated with an air quality modelling case study.
- MOBILE6.2-Mexico and MOVES-Mexico traffic emissions are compared.
- Modelled concentrations of CO, NO_2 and O_3 are improved when using MOVES-Mexico.
- O_3 peaks are increased in the core urban area when reducing traffic emissions.

GRAPHICAL ABSTRACT



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ABSTRACT

This article describes the High-Elective Resolution Modelling Emission System for Mexico (HERMES-Mex) model, an emission processing tool developed to transform the official Mexico City Metropolitan Area (MCMA) emission inventory into hourly, gridded (up to 1 km^2) and speciated emissions used to drive mesoscale air quality simulations with the Community Multi-scale Air Quality (CMAQ) model. The methods and ancillary information used for the spatial and temporal disaggregation and speciation of the emissions are presented and discussed. The resulting emission system is evaluated, and a case study on CO, NO_2, O_3, VOC and $PM_{2.5}$ concentrations is conducted to demonstrate its applicability. Moreover, resulting traffic emissions from the Mobile Source Emission Factor Model for Mexico (MOBILE6.2-Mexico) and the MOtor Vehicle Emission Simulator for Mexico (MOVES-Mexico) models are integrated in the tool to assess and compare their performance. NO_x and VOC total emissions modelled are reduced by 37% and 26% in the MCMA when replacing MOBILE6.2-Mexico for MOVES-Mexico traffic emissions. In terms of air quality, the system composed by the Weather Research and Forecasting model (WRF) coupled with the HERMES-Mex and CMAQ models properly reproduces the pollutant levels and patterns measured in the MCMA. The system's performance clearly improves in urban stations with a strong influence of traffic sources when applying MOVES-Mexico emissions. Despite reducing estimations of modelled precursor emissions, O_3 peak averages are increased in the MCMA core urban area (up to 30 ppb) when using MOVES-Mexico mobile emissions due to its VOC-limited regime, while concentrations in the surrounding suburban/rural

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areas decrease or increase depending on the meteorological conditions of the day. The results obtained suggest that the HERMES-Mex model can be used to provide model-ready emissions for air quality modelling in the MCMA.

1. Introduction

Emission inventories are recognized as key inputs for air quality modelling, specially when they are used in high-resolution forecast systems (Guevara et al., 2013; Pay et al., 2014) or to design effective control measures to mitigate the adverse impact of air pollution (Baldasano et al., 2014; Soret et al., 2014). At the same time, several studies have identified emission inventories as one of the main sources of uncertainty in the air quality modelling chain (e.g. Russel and Dennis, 2000; Hanna and Davis, 2001). Therefore one of the key aspects of emission inventories is its representativeness in terms of spatial resolution (e.g. Mensink et al., 2008), temporal resolution (e.g. Menut et al., 2012) and chemical disaggregation (e.g. Heo et al., 2012).

Despite the important improvements observed during the last decades (Stephens et al., 2008) air pollution is still a major concern in the Mexico City Metropolitan Area (MCMA) (Retama et al., 2015, Jaimes-Palomera et al., 2016). The concentrations of ozone (O_3) and particles (i.e. PM_{10}, $PM_{2.5}$) still exceed the standard thresholds set up by the Official Mexican Standard (NOM; 95 ppb – 1 h and 70 ppb – 8 h moving average for O_3, 50 $\mu g \cdot m^{-3}$ – annual average for PM_{10} and 15 $\mu g \cdot m^{-3}$ – annual average for $PM_{2.5}$), while the levels of sulphur dioxide (SO_2), carbon monoxide (CO) and, to a lesser extent, nitrogen dioxide (NO_2) have been reduced in recent years (SEDEMA, 2015). Observation-based field campaigns and model-based numerical simulations performed in the framework of the MCMA-2003 (Molina et al., 2007) and MILAGRO 2006 (Molina et al., 2010) projects have allowed improving the understanding of the chemistry, dispersion and transport processes of the pollutants emitted to the MCMA atmosphere.

Starting in 1994 the Mexico City's Secretariat of the Environment (SEDEMA) has been publishing biennial emission inventories for the MCMA. On the other hand, in 2006 the Mexico's National Institute of Ecology (INE) together with the United States Environmental Protection Agency (US EPA) finished the development of a national emissions inventory (NEI) and ancilliary data for Mexico, taking 1999 as the reference year (INE-SEMARNAT, 2006). The following national emission inventories for Mexico (INEM 2005, 2008 and 2013) have been fully developed by the National Atmospheric Emission Inventory System (SiNEA), which is supported by the Secretary of the Environment and Natural Resources (SEMARNAT) (http://sinea.semarnat.gob.mx/sinea.php). The MCMA and INEM emission inventories have been used in the modelling studies developed under the MCMA-2003 and MILAGRO 2006 projects (e.g. Zavala et al., 2009a, b; Li et al., 2011). In all cases, total annual emissions have been converted into spatially and temporally distributed, speciation files appropiated for the specific modelling episodes. Nevertheless, the methodologies and ancillary data used have not been deeply presented or discussed in any case, and the work performed has not been translated in the development of a versatile and flexible tool for processing Mexican emissions for atmospheric models.

Current official emission inventories developed in Mexico for regulatory purposes are still reported as total annual emissions per pollutant sector and administrative unit (i.e. municipality) and they do not match the air quality model requirements (i.e. inputs in the form of grid cell, hourly and chemical species-based emissions). Consequently, a reliable emission processing system capable of creating emission data suitable for air quality modelling in the MCMA is needed. Until now, many tools and software systems have been developed to process official emission inventories in different parts of the world (e.g. Monforti and Pederzoli, 2005; Keller et al., 2014). One example of these emission processing systems is the Sparse Matrix Operator Kernel System (SMOKE), developed by the MCNC Environmental Modelling Center (EMC) to support air quality modelling activities in the United States (Houyoux and Vukovich, 1999) and successfully adapted to other regions including the whole of Europe (Bieser et al., 2011), Spain (Borge et al., 2008) and the Pearl River Delta region (Wang et al., 2011).

As mentioned before, SEDEMA is in charge of developing the official annual emission inventory for the MCMA. The emission inventory is computed using bottom-up methods and emission factors, which are either derived from local measurements or taken from the literature. Until now, MCMA's mobile source emissions have been calculated using the emission factors generated by the Mobile Source Emission Factor Model for Mexico (MOBILE6.2-Mexico) (ERG, 2003). The MOBILE6.2-Mexico emission factor model is based upon US EPA's MOBILE6 model (USEPA, 2003), as well as some location-specific emission factor models previously developed for Mexico's metropolitan areas (ERG, 2000; ERG, 2001a; ERG, 2001b) and other technical studies conducted in the country (Schifter et al., 2003). Despite assembling data from previous local works to adapt the model to Mexico, the emission rates and degradation factors of the MOBILE6.2-Mexico model are based upon a relatively small dataset of emission testing results (< 1000 vehicles) that are currently outdated. Direct measurements of mobile emissions performed during the MILAGRO campaign in the MCMA (Schifter et al., 2008; Zavala et al., 2009a; Thornhill et al., 2010) indicated an overestimation of the 2006 MCMA traffic emissions of 20–28% for CO and 14–20% for NO_x. A more recent study suggested that using the US EPA MOVES2010a emission model instead of the MOBILE6.2-Mexico could increase the $PM_{2.5}$ traffic emissions in the MCMA (Zavala et al., 2013).

Considering all of the above, and due to technological advances in motor vehicles, the National Institute of Ecology and Climate Change (INECC) recently required an update of the emission factor estimation tool to the MOtor Vehicle Emission Simulator for Mexico (MOVES-Mexico) for official emission reporting (INECC, 2014). Like MOBILE6.2-Mexico, MOVES-Mexico is a customization of the original North American MOVES2010b model to reflect local conditions (Yang et al., 2012; ERG, 2016). Mexico emission data collected between 2008 and 2014 using Remote Sensing Devices was used to calibrate the original US default emission rates and deterioration factors (Koupal et al., 2016). MOVES-Mexico also considers Mexican vehicle emission and fuel quality standards.

There have indeed been some comparisons between the performances of the MOBILE6 and MOVES models in the US (e.g. Vallamsundar and Lin, 2011; Rappenglueck et al., 2013). However, to the author's knowledge, no similar comparison or validation studies have been carried out to date in Mexico. Considering that exhaust emissions from on-road transport present a significant contribution to total NO_x (79%), CO (96%), PM_{10} (20%) and $PM_{2.5}$ (28%)in the MCMA (SEDEMA, 2016), the impact of changing the emission factor model on air quality modelled concentrations needs to be assessed.

This manuscript describes the High-Elective Resolution Modelling Emission System for Mexico (HERMES-Mex), a model-ready emission processing system specifically developed for the MCMA. The modelling scheme proposed here couples the two official emission inventories that cover the MCMA with the CMAQ chemistry transport model: the local MCMA inventory, developed by the SEDEMA, and the national INEM inventory developed by SEMARNAT. The methods and ancillary information used for the spatial and temporal disaggregation and speciation of the emissions are presented and discussed. A case study is performed with the aim of: (i) demonstrating the applicability of HERMES-

Mex to provide air quality model-ready high resolution spatial, temporal, and speciation emission inputs, (ii) comparing and evaluating the performance of MOBILE6.2-Mexico and MOVES-Mexico and identifying which model may be more accurate to simulate air quality concentrations in the MCMA and (iii) analyzing the O_3 sensitivity to mobile-source emissions in the MCMA.

The structure of the article is as follows: Section 2 describes the methods and ancillary information used in the HERMES-Mex model as well as the model setup and the observational dataset used for the case study. In Section 3 a comprehensive analysis of the emission results is conducted along with a comparison of the modelled concentrations against available observational data. Finally, Section 4 summarises and discusses the results.

2. Methodology

The following sections present the conceptual framework of the tool, followed by the description of the emission inventories used as input and the methods and ancillary data used to break down total emissions spatially, temporally, and by model specie. Finally, the case study and modelling domains used to test the system are introduced.

2.1. Conceptual framework

HERMES-Mex is a stand-alone software component for converting Mexican official annual emission data from different sources (i.e. area, mobile or point sources) and pollutants (i.e. SO_2, CO, NO_x, VOC, TOC, NH_3, PM_{10}, $PM_{2.5}$ and PM_{coarse},) into the formatted emission input files required by CMAQ. HERMES-Mex acts as a coupler between a set of emission inputs and ancillary data organised in a data library and the external chemical transport model. The emission processing system consists of five modules that are responsible for the horizontal (Section 2.3), vertical (Section 2.4) and temporal (Section 2.5) allocation, chemical speciation (Section 2.6) and writing of the output emission files. Input data is provided to the system using cross-reference text files and a customizable configuration file. The system has the capacity of using as input meteorological information from the WRF model and biogenic emissions estimated by the Model of Emissions of Gases and Aerosols from Nature (MEGANv2.1) (Section 2.2). The development of the model is based on the knowledge acquired from previous versions of HERMES for Europe (Ferreira et al., 2013) and Spain (Guevara et al., 2013). Detailed information on the workflow and data handling of the model can be found in the Supplementary material.

2.2. Emission datasets

Two official inventories of anthropogenic emission are used by the tool: the MCMA, developed by the SEDEMA, and the INEM, developed by the SEMARNAT. The former is applied to all MCMA's municipalities (red area in Fig. 1), while the latter provides emission data to the rest of municipalities covered by D1 and D2 working domains. The main characteristics of both inventories are presented in this section. For a more specific description of the methodologies and data used to estimate the MCMA and INEM inventories, authors refer to SEDEMA (2016) and the corresponding official websites of the emission developers, where several open-access reports are available (SEDEMA, http://www.aire.cdmx.gob.mx; SEMARNAT, http://sinea.semarnat.gob.mx/sinea.php).

The two inventories report annual emissions for NO_x, VOC, COT, CO, SO_2, NH_3, PM_{10} and $PM_{2.5}$ and cover all major emission sources, including a total of 81 general sectors divided into: point sources (23), area sources (45) and mobile sources (13). Point source emissions include information on the industrial sector (i.e. energy, pulp, paper and print, food processing, beverages and tobacco, chemicals, non-metallic minerals, iron and steel, non-ferrous metals, wood processing, plastics, textiles and other manufacturing industries), geographical location and stack properties for each individual facility. Emissions from area and mobile sources are reported at the municipality level. If combustion processes are involved, they are classified according to the burned fuel (area and mobile sources) and type of vehicle (mobile sources). Both MCMA and INEM report the emissions following the same format and subsequently no harmonization is required when the two datasets are implemented in the model. In the present work the most recent versions of both inventories are used, which correspond to the years 2014 (MCMA) and 2013 (INEM).

Point source emissions are compiled using the information provided by the Annual Operations Certificate (COA) of each installation, which includes information of the fuel type and fuel consumption, energy consumption, type of equipment used (e.g. turbine, boiler), stack parameters (e.g. stack height, exit gas temperature and velocity) and annual emissions released to the atmosphere, among others. In the case of the MCMA inventory, the information reported by the COAs passes a quality control based on local expertise (i.e. emissions are reallocated and recalculated if needed) and the point source database is complemented with the data obtained from Unique Environmental Licences (LAU), which include small and medium facilities not contemplated in the COAs. Currently a total of 5197 point sources are considered in the MCMA inventory.

Emissions from area sources are computed using local activity data provided by different organizations (e.g. Secretariat of Energy, SENER; National Institute for Statistics, Geography and Information Technology, SINEGI) and emission factors developed by the US EPA (https://www3.epa.gov/ttnchie1/ap42/) or studies conducted in Mexico for specific local sources (e.g. LGP distribution and leakage emissions; Gamas et al., 2000). Specific emission models are used for certain categories such as Tanks v4.0.9D (https://www3.epa.gov/ttn/chief/ap42/ch07/) for fuel storage tanks evaporation.

Mobile source emissions are estimated combining a set of traffic input data (e.g. vehicle population by age, vehicle type and fuel type, miles travelled by year) and one of the emission models mentioned above. The fleet characterization was obtained from the Inspection and Maintenance (I/M) Program database (SEDEMA, personal communication), which is complemented with the federal fleet and public transport databases reported by the Secretariat of Communications and Transport (SCT, www.gob.mx/sct) and the Secretariat of Mobility (SEMOVI, www.semovi.cdmx.gob.mx/), respectively. In the present work two versions of the MCMA emission inventory are considered; one using the MOBILE6.2-Mexico model and another one using MOVES-Mexico (see Section 1 and Table 1 from the Supplementary material). In the case of the INEM inventory, only the version that uses MOVES2010b is available. Nevertheless, it is expected that its influence on the MCMA modelled pollutant concentrations will be negligible due to two facts: (i) Within the areas where the INEM inventory is used, the municipality with the highest population density (Toluca) is located ~25 km away from the MCMA and (ii) the total mobile emissions estimated for this municipality are ~15 times lower than those obtained for the entire MCMA.

For biogenic emissions, the emission processing tool integrates the MEGANv2.1 model (Guenther et al., 2012) to estimate gridded, hourly, and speciated VOC, CO and NO natural emissions. The model uses meteorological inputs from WRF (i.e. temperature, solar radiation and soil moisture) and the official landcover datasets that are available online (http://lar.wsu.edu/megan/).

2.3. Spatial distribution

Spatial surrogates are used to allocate the reported municipal emissions to the working model grid. The tool uses the classical approach of computing activity ratios for each grid cell based on spatial proxies that have been previously referenced to the grid using a Geographical Information System (GIS) (Dai and Rocke, 2000). Apart from the spatial proxies, information on the fraction of the municipality/ies that

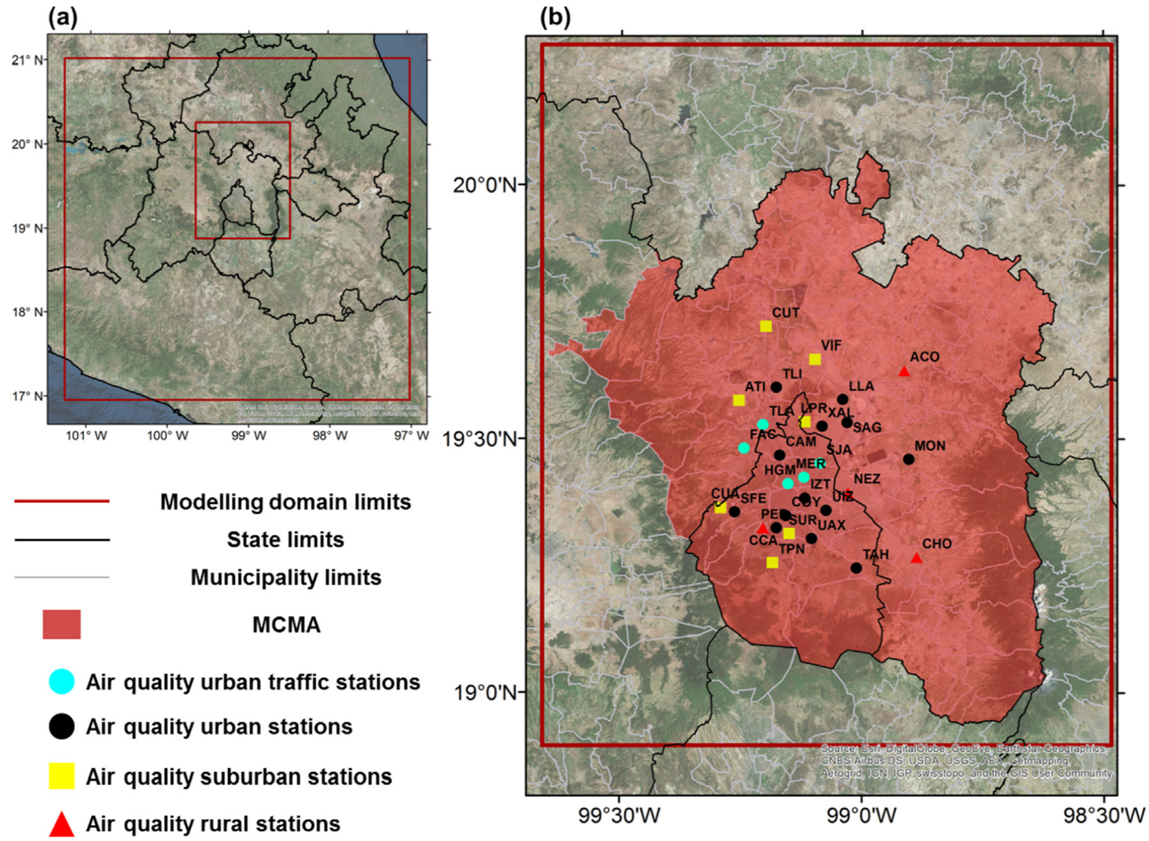

Fig. 1. Meteorology. emission and air quality modelling domains D1 (3 km by 3 km) (a) and D2 (1 km by 1 km) (b) (outer red square). The area shaded in red corresponds to the MCMA. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

intersect with each grid cell (from 0 to 1) is also used to compute these activity ratios.

In order to avoid losing the bulk emissions, the tool includes a checking process that ensures that the sum of all the activity ratios for a given surrogate across the grid cells of a given municipality is equal to one. Moreover, the emission tool estimates and compares total annual emissions before and after the spatial allocation process to verify that input data has been correctly processed.

Table 1 lists the spatial proxies used to allocate the emissions from area sources. For each type of source, up to three different spatial proxies are proposed. HERMES-Mex uses by default the first option assigned to each source category. If none of the cells that intersect the corresponding municipality have information of the selected proxy, the next spatial surrogate is automatically selected. A mass conservative area interpolation (i.e. area cell balance) is applied to the emissions when none of the proposed spatial proxies is available. This process allows avoiding the loss of input emission when the proposed spatial surrogate is not available in a certain municipality. Note that 86% of the total emissions are allocated by the 1st spatial proxy, 9% by the 2nd and 5% by the 3rd.

Urban and agricultural land use information is obtained from the European Space Agency (ESA) Climate Change Iniciative Land Cover (CCI-LC) 300 m data (http://maps.elie.ucl.ac.be/CCI/viewer/index.php), which uses the Medium Resolution Imaging Spectometer (MERIS) Full and Reduced Resolution (FR and RR) and SPOT-Vegetation archives to produce a 5-year epoch (2010, 2008–2012) global land cover map. Industrial land- use information is obtained from the Open StreetMap project (OSM) (Ramm et al., 2011) through the Geofabrik services (https://www.geofabrik.de) and has been manually completed according to the location of industrial facilities reported by the MCMA emission inventory. The population density is discriminated between urban and rural categories and it is obtained from the National Institute for Statistics, Geography and Information Technology (SINEGI). In the case of the MCMA, population data are taken from the AGEBS (Basic Geostatistical Areas), which are the smallest levels of geography (after census-blocks) at which Mexico's Census tabulates demographic data. Emissions from airports, bus terminals and solid waste disposal are assigned to the corresponding infrastructures, which have been digitalised using a Geographic Information System (GIS) and Environmental Systems Research Institute's (ESRI) satellite imagery data. For certain source categories (e.g. waste-water handling, gasoline distribution) information on the location of the corresponding installations where the activity is performed is used (e.g. wastewater treatment plants, gas stations). A weight factor is assigned to each installation depending on the amount of activity load (e.g. amount of water treated in each wastewater treatment plant).

A digitalised road network map obtained from OSM is combined with traffic flow information per vehicle type derived from MCMA traffic counts (ETEISA, 2005) to generate the spatial surrogate for mobile sources. The road network map is classified according to eight different types of roads, including: motorway, trunk road, primary road, secondary road, tertiary road and residential road. The traffic flow information is used to assign specific weight factors to each type of road and vehicle (Table 2). This allows a more realistic distribution of mobile emissions throughout the whole road transport infrastructure (e.g. circulation of heavy duty vehicles is more restricted to motorways while passenger cars also circulate through residential roads). For the case of the MCMA Bus Rapid Transit (BRT), the official digitalised bus lane is used, whereas in the case of unpaved resuspension emissions, a specific unmade road network map is applied (SIGSA, 2002). Considering that congestion plays a very important role in traffic emissions (e.g. Quaassdorff et al., 2016), future works will explore the possibility of including average speed in the surrogate generation process (besides traffic volume).

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Table 1
List of spatial surrogates used to spatially allocated area emissions.

Emission Source	Spatial proxy (1)	Spatial proxy (2)	Spatial proxy (3)
1. Industrial combustion (LGP, diesel, natural gas)	Industrial land uses	Urban land uses	Urban population
2. Commercial combustion (LPG, natural gas)	Urban population	Urban land uses	Area cell balance
3. Residential combustion (LPG, natural gas)	Urban population	Rural population	Area cell balance
4. Residential combustion (wood, querosene)	Rural population	Urban population	Area cell balance
5. Agricultural combustion (LPG, diesel, querosene)	Agricultural land uses	Area cell balance	
6. Food and drink (bakeries)	Urban population	Urban land uses	Area cell balance
7. Cooking activities (charkool)	Urban population	Urban land uses	Area cell balance
8. Hospital sterilizers	Hospitals	Urban land uses	Area cell balance
9. Construction activities	Urban population	Urban land uses	Area cell balance
10. Road paving with asphalt	Urban population	Urban land uses	Area cell balance
11. Bus terminals	Bus terminals	Urban land uses	Area cell balance
12. Airport activities (international and national aviation, storage and handling of jet fuel)	Airport terminals	Area cell balance	
13. Use of solvents (dry cleaning, printing, industrial and other coating applications, commercial and domestic solvent use, paint applications, adhesives and sealants, miscellaneous products)	Urban population	Urban land uses	Area cell balance
14. LGP distribution, storage and leakage	Urban population	Urban land uses	Area cell balance
15. Gasoline distribution	Gas stations	Urban land uses	Area cell balance
16. Gasoline storage	Fuel storage terminals	Urban land uses	Area cell balance
17. Agriculture (off-road vehicles, fertilizers, pesticides, farming and agricultural harvest, burning of agricultural waste)	Agricultural land uses	Area cell balance	
18. Cattle raising	Agricultural land uses	Area cell balance	
19. Waste-water handling	Waste-water treatment plants	Urban land uses	Area cell balance
20. Solid waste disposal on land	Landfills	Urban land uses	Area cell balance
21. Composting	Composting plants	Urban land uses	Area cell balance

In the case of point sources, for which geographic latitude and longitude is available, emissions are directly allocated to the grid cells that intersect with each facility.

The Supplementary material provides graphical representations of the different spatial proxies described in the present section (Supplementary material, Figs. 2 and 3).

2.4. Vertical distribution

Once the system has horizontally allocated the total annual emissions, the next step consists of vertically distributing them across the vertical layers of the 3D modelling domain (Section 2.7).

Table 2
Weight factors assigned to each type of vehicle category and road (Mot: motorway; Tru: trunk road; Pri: primary road; Sec: secondary road; Ter: tertiary road, Res: residential road, Bus: bus lane and Unm: unmade road).

Mobile source category	Weight factors by road type (from 0 to 1)							
	Mot	Tru	Pri	Sec	Ter	Res	Bus	Unm
Passenger cars and SUV (gasoline, diesel, natural gas, LPG)	0.198	0.198	0.217	0.202	0.138	0.046	0.000	0.000
Taxis (gasoline, diesel, natural gas, LPG)	0.142	0.251	0.243	0.189	0.132	0.044	0.000	0.000
Buses and microbuses (gasoline, diesel, natural gas, LPG)	0.217	0.313	0.192	0.129	0.150	0.000	0.000	0.000
Combis (gasoline, diesel, natural gas, LPG, hybrid)	0.148	0.262	0.172	0.202	0.162	0.054	0.000	0.000
Pickup truck and light duty vehicles gasoline, diesel, natural gas, LPG)	0.140	0.205	0.190	0.181	0.214	0.071	0.000	0.000
Heavy duty vehicles (gasoline, diesel, natural gas, LPG)	0.423	0.200	0.149	0.228	0.000	0.000	0.000	0.000
Tractor units (diesel, gasoline)	0.423	0.200	0.149	0.228	0.000	0.000	0.000	0.000
Motorcycles (gasoline)	0.142	0.251	0.243	0.189	0.132	0.044	0.000	0.000
Metrobus and Mexibus (diesel)	0.000	0.000	0.000	0.000	0.000	0.000	1.000	0.000
Resuspension (paved road)	0.198	0.198	0.217	0.202	0.138	0.046	0.000	0.000
Resuspension (unpaved road)	0.000	0.000	0.000	0.000	0.000	0.000	0.000	1.000

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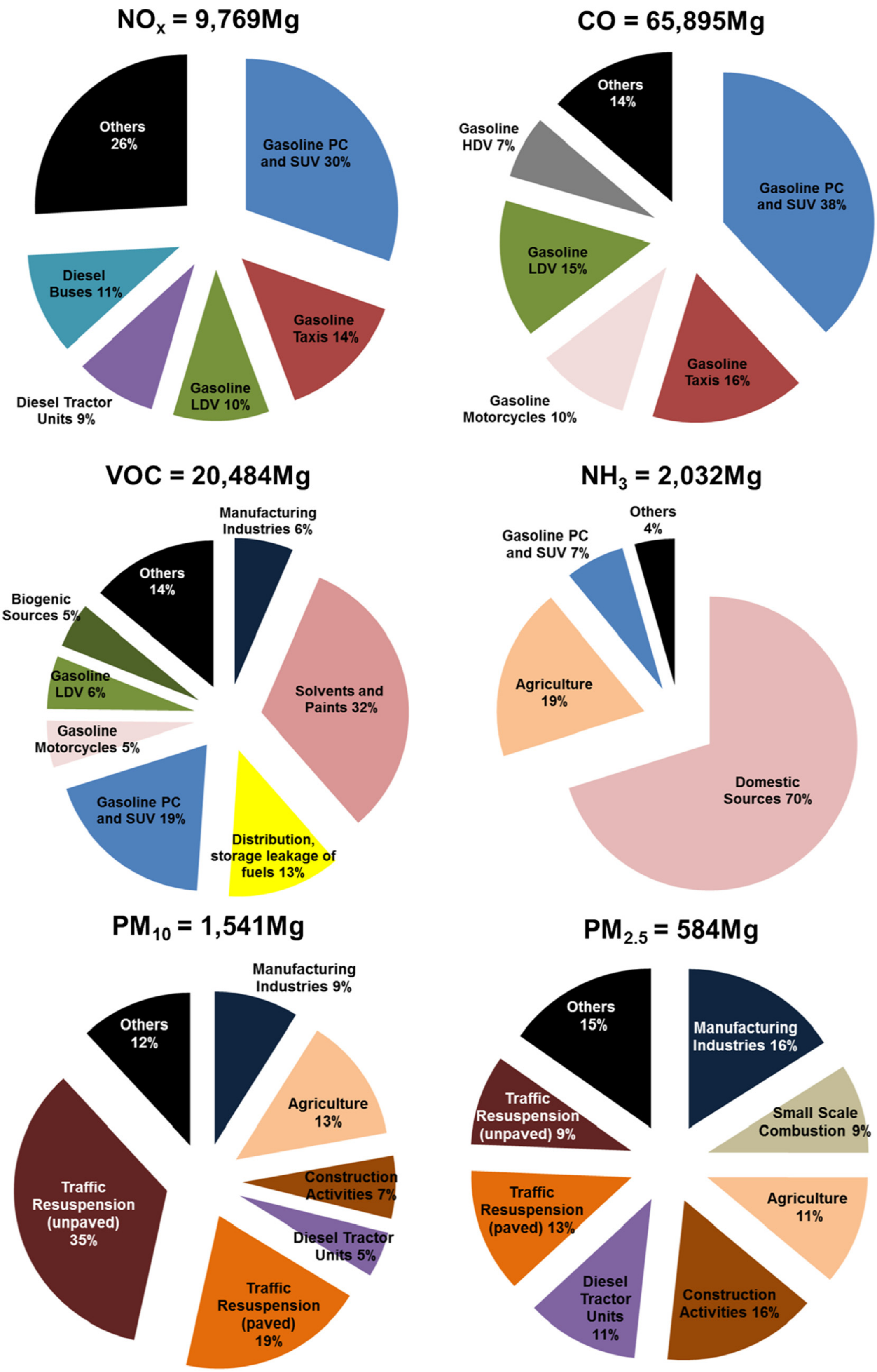

Fig. 2. Sector contribution for NO_x, CO, VOC, NH_3, PM_{10} and $PM_{2.5}$ total emissions in the MCMA during the case study (14–28 February 2014).

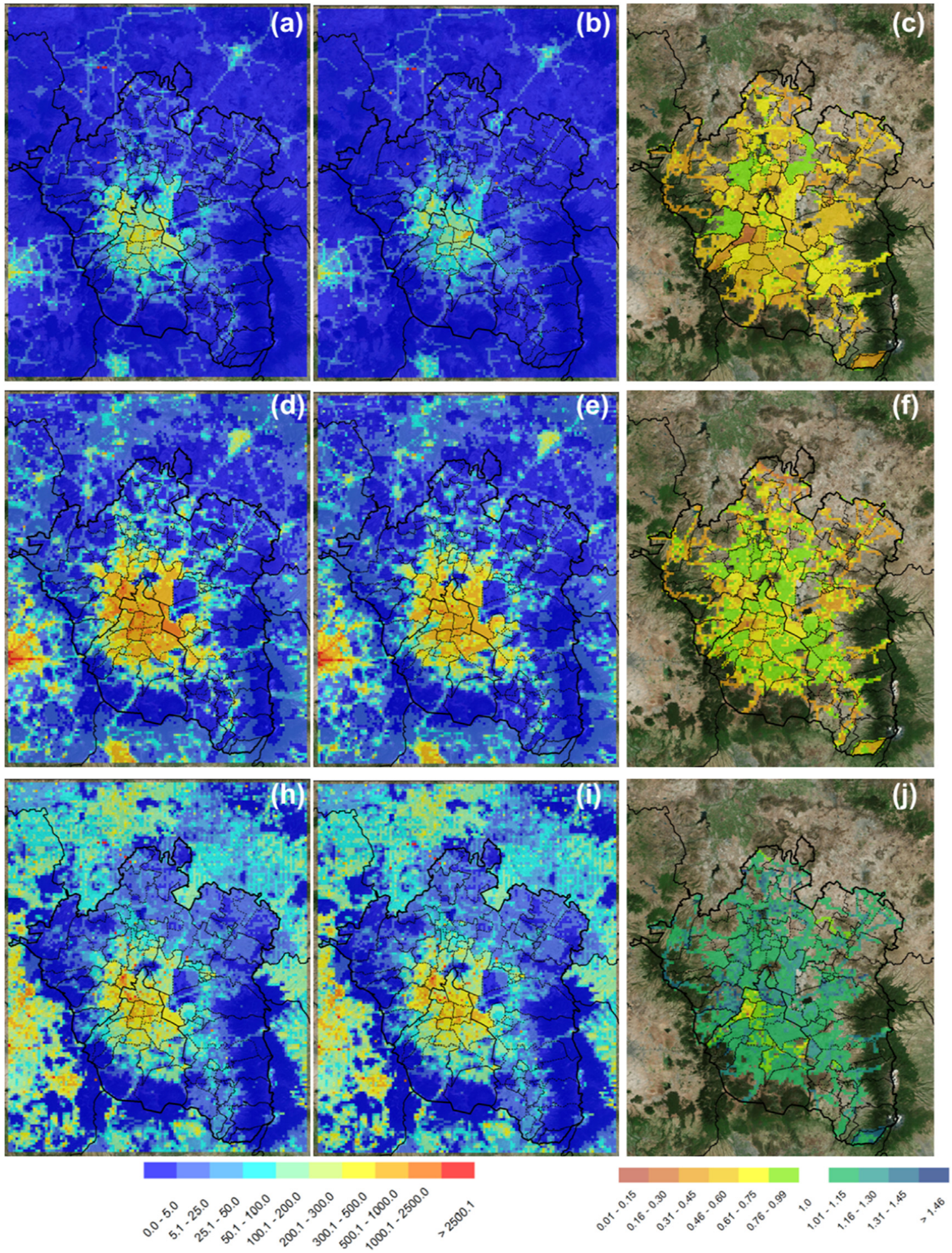

Fig. 3. Spatial distribution of NO_x (a to c), VOC (d to f) [$mol \cdot km^{-2}$] and $PM_{2.5}$ (h to j) [$g \cdot km^{-2}$] emissions in the MCMA for the period of study when applying MOBILE6.2-Mexico (a, d and h) MOVES-Mexico (b, e and i) and the difference between results (c, f and j).

The system assumes that area and mobile source emissions are released into the lowest vertical layer, whereas in the case of point source emissions their vertical allocation is performed using fixed vertical profiles based on the stack height of each facility (Guevara et al., 2014a).

2.5. Temporal distribution

Hourly distribution of annual emission estimates is performed through the application of 21 monthly profiles, 16 weekly profiles and 121 hourly profiles.

The temporal variations of area and point source emissions are obtained from source-specific temporal profiles collected by SEDEMA. Point source profiles are based on the working hours information reported by the COAs and are classified according to the different industrial sectors. The temporal distributions of area emissions are based on industrials profiles (e.g. graphic arts, chemicals, food processing), electricity demand and population schedules (residential and commercial sources), and travel itineraries (non-road transport). In the case of mobile sources, temporal profiles are obtained from hourly traffic counts registered by a total of 206 traffic stations in the MCMA during the year 2015 (Secretariat of Works and Services of Mexico City, personal communication). Specific profiles are applied to each Mexico City's municipality with available information, while in the rest of the domain the resulting average profiles are used. Hourly profiles are distinguished between working days, Saturdays and Sundays to allow a more realistic temporal disaggregation.

The effect of rain on the PM emissions from road dust resuspension is considered by the model through the implementation of the recovery expression reported by Amato et al., (2012). The formula, which is based on measurements undertaken in Barcelona (Spain) and Utrecht (the Netherlands), indicates that after a rainfall (when the mobility particles drops to values close to zero), the loading of mobile road dust mobility increases exponentially tending to reach again the maximum emission strength. The equation depends on a recovery rate that varies according to the traffic characteristics and local climatic conditions. Since no local measurements are available for Mexico, the Barcelona recovery rate is applied due to its similarity (average annual temperature and relative humidity in Barcelona and Mexico are 15.5 °C and 17.0 °C and 72% and 57.6%, respectively). Mobile road dust recovery reaches 50% and 75% after 8 and 16 h, and the total recovery occurs after 48 h. Precipitation information is obtained from WRF and the temporal correction is applied when at least a 0.254 mm rainfall occurs (Supplementary material, Fig. 4).

2.6. Chemical speciation

Source-specific speciation profiles for VOC and $PM_{2.5}$ emissions are developed for the Carbon Bond 05 (CB05) chemical mechanism (Yarwood et al., 2005) and the AERO5 module based mainly on the SPECIATE4.4 database (US EPA, 2014). VOC emissions from gasoline and diesel mobile sources, handling and distribution of LPG, asphalt operations, painting operations and landfills are speciated according to local measurements performed in the MCMA by Mugica et al. (2001 and 2002). $PM_{2.5}$ emissions released by unpaved and paved road resuspension and agricultural harvest are obtained from Vega et al. (2001).

For the NO_x speciation, a constant NO/NO_x ratio of 0.9 is assumed for all the area and point source categories, following US EPA (2014). In the case of mobile sources, the NO/NO_x and NO_2/NO_x ratios vary according to the vehicle category. While gasoline-powered cars are given a constant NO_2/NO_x ratio of 0.04, diesel-powered vehicles present values

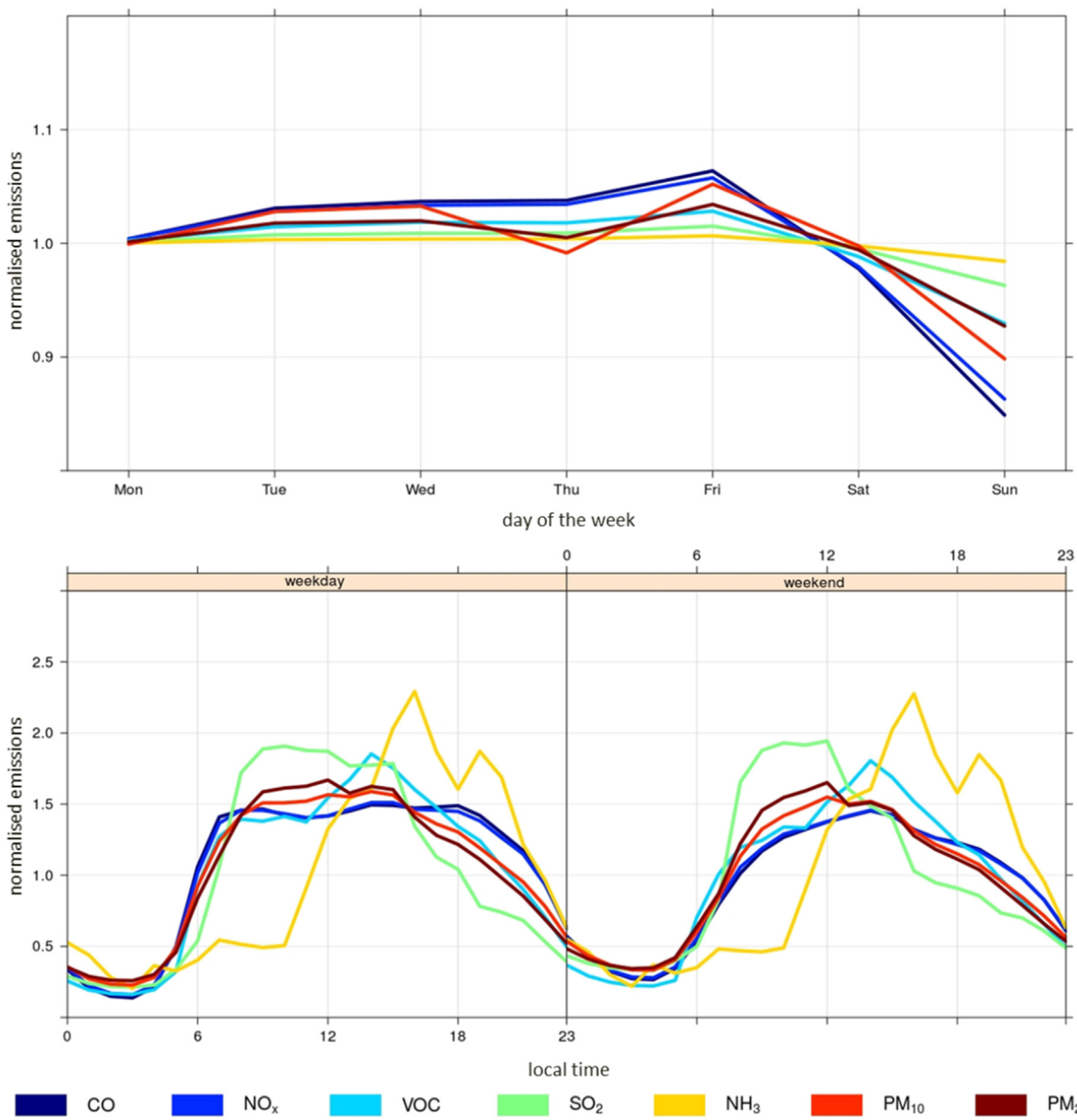

Fig. 4. Weekly (top) and daily (bottom) variations in CO, NO_x, VOC, SO_2, NH_3, PM_{10} and $PM_{2.5}$ on weekdays and weekends for the period of study (14–28 February 2014).

that range from 0.14 (diesel trucks) up to 0.30 (diesel passenger cars) (Carslaw and Rhys-Tylerb, 2013). Nitrous acid (HONO) is also considered in the speciation of traffic NO_x emissions following the recommendations by Li et al. (2010), which indicated that HONO sources are found to significantly improve O_3 and secondary aerosol concentrations when performing air quality simulation studies over the MCMA. Therefore, a 0.008 HONO/NO_x ratio is applied to emissions from road transport (Kurtenbach et al., 2001). The HONO fraction is subtracted from the NO_2 fraction such that all fractions sum one (i.e. NO, NO_2 and HONO).

For biogenic emission, the conversion table reported by MEGANv2.1 to assign the 150 biogenic VOC species to the CB05 chemical mechanism species is used.

2.7. Case study: model configuration and observational data

A case study using the WRF meteorological model (Skamarock and Klemp, 2008), HERMES-Mex and the CMAQ chemical transport model (Byun and Schere, 2006) is performed to evaluate the emission processing system and analyze the performance of MOBILE6.2-Mexico and MOVES-Mexico in terms of air quality. The simulation period for the case study is from February 14 to February 28, 2014. During this period an episode of O_3 pollution affected the MCMA. The hourly limit value set up by the NOM-020-SSA1-1993 (i.e. 110 ppb) was exceeded in 22 air quality stations and the environmental pre-contingency alert for O_3 was activated (SEDEMA, 2015). It is important to note that since January of 2015 the hourly limit value for O_3 has been reduced from 110 ppb to 95 ppb (NOM-020-SSA1-2014). Nevertheless, and since this simulation is performed for the year 2014, the limit value of 110 ppb is considered during the present study. A spin-up period of 3 days (i.e. February 11–13) was used to minimize the influence of the initial conditions.

The system is built over the two nested domains (D1: 3 × 3 km and D2: 1 × 1 km) (Fig. 1) in which HERMES-Mex is currently configured. D1 consists of 151 × 151 grid cells and covers the central region of Mexico, while D2 is centred at −99.0° lon and 19.6° lat and encompasses the whole MCMA region. Both domains are configured with a total of 37 sigma levels with top pressures of 50 hPa. Note that since HERMES-Mex uses the national INEM inventory, the dimensions and location of the HERMES-Mex mother domain are customizable as long as the covered area is restricted to Mexican territory. Nevertheless, and in order to correctly execute the model in a different domain, users should redefine all the input ancillary data that depends on the grid definition (i.e. horizontal allocation files, see Fig. 1 from the Supplementary material).

WRF version 3.6.1 is used to generate the meteorological fields required by the chemical transport model. An evaluation of WRF for the case study shows that the model is capable of reproducing the diurnal cycle of the observations in terms of temperature, relative humidity, wind speed and wind direction. Cold biases on the order of 2–2.5 °C are observed during daytime, which implies a daytime wet biases of the relative humidity (~5.2%). On the other hand, significant overestimations of wind speed are observed during the afternoon (~2.5 $m \cdot s^{-1}$), when the wind speed reaches its maximum value. Details about the configuration and the performance of WRF for the case study are provided as Supplementary material (Supplementary material, Figs. 6–9).

The boundary conditions for the coarsest domain in CMAQ version 5.0.2 model are obtained from the Model of Ozone and Related Tracers (MOZART-4; Emmons et al., 2010) data available at every 6 h. MOZART-4 species are converted to CMAQ CB05 species following the mapping proposed by Tai et al. (2008). Boundary conditions for the inner domain are generated from the D1 domain. For both domains, initial conditions are generated using the default profiles in the CMAQ model. The gas-phase chemistry mechanism used is the CB05TUCL (Whitten et al., 2010). The aerosols are modelled using the AERO5 module (Binkowski and Shankar, 1995). The Asymmetric Convective Model version 2 (ACM2) (Pleim, 2007) is used to calculate vertical diffusion and the horizontal and vertical advection and modelled using the piecewise parabolic method (PPM) (Colella and Woodward, 1984). The Meteorology Chemistry Interface Processor (MCIP) (Otte and Pleim, 2010) is used to reformat the WRF output into the CMAQ-ready input data.

In order to evaluate the performance of the emission system by means of air quality (i.e. CO, NO_2, O_3, $PM_{2.5}$), a comparison with measurements from the Red Automática de Monitoreo Atmosférico (RAMA) (http://www.aire.df.gob.mx/) is performed. The RAMA air quality monitoring network is supported by SEDEMA and collects hourly records of ambient concentrations of the so called criteria pollutants (i.e. CO, NO_2, O_3, SO_2, PM_{10}, $PM_{2.5}$) from 34 monitoring stations (28 in the year 2014) (Fig. 1). For this work, all the stations with a temporal coverage below 80% of the entire period were discarded. Stations have been classified according to the surrounding environment (i.e. urban, URB; suburban, SUB; rural, RUR), giving a special focus on those located in areas with a strong influence of traffic sources (URB-TRF). The evaluation also includes the comparison of modelled and observed VOC to CO ratios using special field study VOC samples collected at two RAMA stations (i.e. La Merced, MER and Pedregal, PED, Fig. 1). Despite having a 2 year difference between the study period and the time when the ambient measurements were taken (i.e. 2012) results may be relevant to asses the accuracy of the VOC emissions. Details on the methodolgy performed to collect the sample can be found in Jaimes-Palomera et al. (2016). The openair R package (Carslaw and Ropkins, 2012) is used to perform the statistical analysis of the air pollution data.

3. Results

The following sections discuss and evaluate the results obtained in terms of emissions and modelled concentrations.

3.1. Emission results

3.1.1. Total emissions per sector

The contribution of main sources to total emissions in the MCMA during the case study is shown in Fig. 2. The results are estimated using the MOBILE6.2-Mexico traffic emissions.

Gasoline-powered vehicles dominate the total burden of NO_x (~60%) and CO (~92%) emissions, passenger cars (PC) and sport utility vehicles (SUV) being the main contributors with fractions of 30% (NO_x) and 38% (CO). Diesel vehicles (i.e. HDV, tractor units and buses) also represent an important source of NO_x (~25%) although the amount of vehicle kilometres travelled (VKT) of these vehicle categories is only a 4% of the total annual VKT in the MCMA. The use of solvents and paints and the distribution, storage and leakage of fuels (i.e. LPG and gasoline) are together the largest source of VOC emissions (~45%), with gasoline-powered vehicles contributing 36% to this pollutant. The low contribution of biogenic sources (5%) indicates the high degree of urbanization of the MCMA. NH_3 emissions are mainly dominated by miscellaneous domestic sources (e.g. excreta from domestic animals, use of domestic pesticides, emissions from the sewer system, 70%) and agricultural activities (i.e. use of fertilizers and farming, 19%). Dust resuspension from unpaved (35%) and paved roads (20%) represents more than half of total PM_{10} emissions, followed by agricultural activities (e.g. tillage and harvesting, 13%), manufacturing industries (e.g. chemical, metallurgy, energy and food industries, 9%) and construction activities (7%). Contribution of traffic resuspension dramatically drops when analyzing $PM_{2.5}$ (22%), especially for unpaved roads (9%) since the dust released by this source is mainly related to the PM coarse fraction (e.g. Denier van der Gon et al., 2013). Diesel-powered vehicles represent a fraction of $PM_{2.5}$ of ~17%, which is dominated by the contribution of diesel tractor units (i.e. heavy duty tow trucks) (11%). Manufacturing industries (16%) and construction activities (16%) are the third largest sources of $PM_{2.5}$. In the case of SO_2 >60% of the emissions are derived from manufacturing industries (not shown).

NO_x, CO and VOC mobile source total emissions modelled are reduced by −42%, −53% and −63% in the MCMA when applying

MOVES-Mexico instead of MOBILE6.2-Mexico (Table 3). Gasoline taxis present the highest reduction of these pollutant emissions (between −80% and −90%). On the other hand, total traffic PM_{10} and $PM_{2.5}$ are increased by +70% and +29%, respectively. In this case, the large increases of emissions from Gasoline PC and SUVs (~+180%) are compensated by the decreases of emissions from Diesel Tractor Units (−40%). When comparing the total emissions in the MCMA, the reductions are similar for NO_x (−37%) and CO (−52%) since traffic is the dominant source (Fig. 2). However, the changes for VOC, PM_{10} and $PM_{2.5}$ are much lower (−26%, +8%, +6%) due to the large contributions of solvent and traffic resuspension emissions to these pollutants.

As summarised in Table 1 of the Supplementary material, there are several features of MOBILE6.2-Mexico and MOVES-Mexico that may be causing the difference between computed mobile sources. The general reduction of NO_x, CO and VOC emissions is mainly due to the updating of the emission factors (based on >250,000 remote sensing device measurements) and the inclusion of the Tier emission standards in the vehicle categories of MOVES-Mexico. The increase of PM_{10} and $PM_{2.5}$ emissions from PC and LDV is due to an updating of the algorithm used to calculate the emission rates for these pollutants. On the other hand, diesel HDV and diesel tractor units present a decrease in PM_{10} and $PM_{2.5}$ since the emission rates used in MOBILE6.2-Mexico were computed assuming a fuel sulphur content of 350 ppm, while the current Mexican law on fuel specifications (i.e. NOM-086-SEMARNAT-SENER-SCFI-2005) limit the sulphur content to 15 ppm from September 2009 (previous limit was 500 ppm).

3.1.2. Spatial distribution

Fig. 3 shows the spatial distributions of NO_x (Figs. 3.a, b, c), VOC (Figs. 3.d, e, f) and $PM_{2.5}$ (Figs. 3.g, h, i) from total emission sources during the case study. Left-column and mid-column show the results when using MOBILE6.2-Mexico and MOVES-Mexico, respectively, while right-column presents the differences (calculated as the quotient between MOVES-Mexico and MOBILE6.2-Mexico emissions).

NO_x emissions are more intense in Mexico City downtown and surrounding districts where there are heavily trafficked transportation networks. Clear road network patterns can be observed in the different accesses to the MCMA, especially in the rural areas. The decline of NO_x when replacing MOBILE6.2-Mexico by MOVES-Mexico traffic emissions is fairly constant in most of the municipalities (decrease by a factor of ~0.23). The different degrees of variations in certain regions are due to multiple facts, including: (i) the vehicle fleet compositions that characterize each region and (ii) the contribution of mobile sources in them.

VOC emissions are concentrated where there is high population density, the background levels observed in the outlying areas being related to biogenic sources. The reduction of VOC when replacing traffic emissions is in general less pronounced than in the case of NOx (factor of ~0.9) due to the major contribution of other sources distinct from traffic (the use of solvents and leakage of LPG). The most significant decrease occurs in downtown Mexico City due to the higher presence of traffic activity.

Table 3
Relative change (%) between NO_x, CO, VOC, PM_{10} and $PM_{2.5}$ emissions when using MOVES-Mexico and MOBILE6.2-Mexico.

	NO_x	CO	VOC	PM_{10}	$PM_{2.5}$
Mobile sources	−42%	−53%	−63%	70%	29%
Gasoline PC and SUV	*−39%*	*−39%*	*−55%*	*196%*	*162%*
Gasoline taxis	*−90%*	*−81%*	*−81%*	*117%*	*18%*
Gasoline motorcycles	*−48%*	*−43%*	*−78%*	*24%*	*9%*
Gasoline LDV	*−54%*	*−58%*	*−71%*	*265%*	*285%*
Gasoline HDV	*−32%*	*−41%*	*−65%*	*163%*	*40%*
Diesel HDV	*−16%*	*−78%*	*−69%*	*−1%*	*−17%*
Diesel tractor units	*−14%*	*−83%*	*−85%*	*−33%*	*−42%*
Diesel buses	*−36%*	*−82%*	*−83%*	*329%*	*284%*
Total sources	−37%	−52%	−26%	8%	6%

$PM_{2.5}$ emissions are distributed not only over the highly populated and trafficked areas but also in suburban/rural areas due to the large contributions of unpaved road dust resuspension, agriculture activities and residential wood combustion. As in the case of NO_x, major point sources are seen throughout the whole domain. Background $PM_{2.5}$ emissions are larger in the municipalities located outside of the MCMA. This is mainly due to a discrepancy between the INEM and MCMA inventories in terms of agricultural waste burning emissions. For this pollutant sector, the total $PM_{2.5}$ emissions reported in the MCMA by the national INEM inventory is >10 times higher than the ones reported by the local MCMA inventory. Inconsistencies between local and national emission inventories have been largely reported in other parts of the world (e.g. Guevara et al., 2014b) and are usually related to discrepancies in terms of activity data and/or emission factors (Guevara et al., 2016; Thunis et al., 2016). This analysis shows how HERMES-Mex may be a useful tool to analyze and understand potential ways to improve the consistency between the MCMA and INEM inventories and find options for inventory harmonization (Vedrenne et al., 2016). When applying MOVES-Mexico $PM_{2.5}$ emissions are slightly increased in a homogeneous way throughout the MCMA region (by a factor of ~1.10). A decrease of up to a factor of 0.70 can be observed in some specific regions due to the higher presence of diesel HDV and tractor units.

3.1.3. Temporal distribution

Fig. 4 shows the weekly and daily variations of major pollutant emissions for the two-week simulation period.

There is an obvious weekday-weekend contrast for NO_x, CO and PM_{10} due to the reduced traffic flow during the weekend. For $PM_{2.5}$ and VOC emissions, this reduction is lower due to the contribution of manufacturing industries/construction activities and use of solvents and paints on Saturdays. Both activities are defined with a flat weekly profile. On the other hand, SO_2 and NH_3 emissions present almost a constant profile. The reduction of emissions observed for PM_{10} and, to a lesser extent for $PM_{2.5}$, on Thursdays is due to several rainfalls that occurred in the MCMA on the 27th February, which affected the traffic resuspension emissions (Supplementary material, Fig. 4).

The daily evolutions of NO_x and CO are greatly influenced by daily variations in traffic flow on the weekdays and weekends. During weekdays, maximum NO_x and CO values are reached between 08:00 and 09:00 Local Time (LT, UTC-6h) and emissions remain fairly constant until 18:00-19:00 LT, when the decrease in traffic activity begins. On the other hand, weekend NO_x and CO emissions present less pronounced rush-hour peaks. This is explained by the drop of morning work trips on Saturdays and Sundays. In the case of PM (especially $PM_{2.5}$), the influence of manufacturing industries is observed both on weekdays and weekends, the industrial activity being particularly intense between 08:00 and 16:00 LT during the weekday and 08:00 and 12:00 LT during the weekend. This activity pattern of the industrial activity is also represented in SO_2 emissions. VOC emissions present a peak at 14:00 LT due to the higher intensity of solvent and paint use in the residential/commercial sectors. Finally, NH_3 mainly follows the pattern of miscellaneous domestic sources (e.g. use of domestic pesticides, excreta from domestic animals).

3.1.4. Chemical speciation

Fig. 5 and Table 4 show the speciation results of VOC and $PM_{2.5}$ primary emissions in the MCMA during the period of study.

The largest proportion of VOCs consists of Paraffin (PAR, 66.4%), followed by nonreactive carbon UNR, 18.6%), which consist on nonreactive carbon atoms, as well as Ethanol (ETOH, 3%) and, to a minor extent, Toluene (TOL, 2.7%). Emissions of PAR are mainly associated with area (50.5%) and mobiles sources (47.0%), specifically leakage, distribution and unburned hydrocarbons from LPG (15%), residential use of solvent products (12%) and gasoline-powered vehicles (41%), respectively (not shown). In the case of TOL and ETOH the main contributors are area

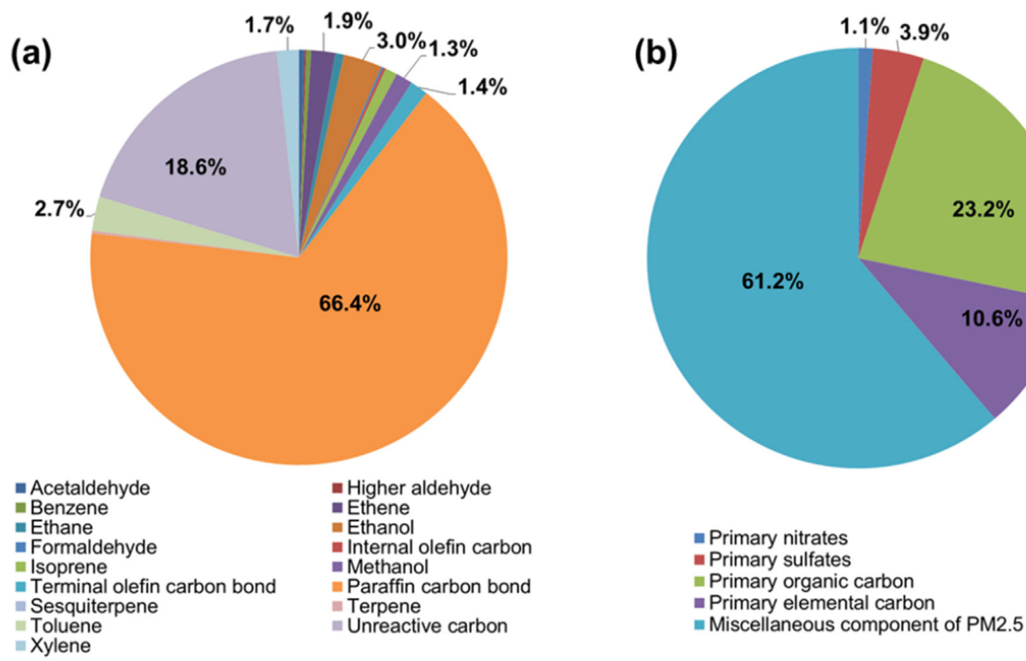

Fig. 5. Chemical components of VOC (a) and $PM_{2.5}$ (b) emissions in the MCMA for the case study.

sources (62.9% and 84.37%, respectively), including architectural use of paint (39%) and the use of home and personal care products (47%), respectively (not shown). Mobile sources present the highest contribution to Benzene (BENZENE, 79.8%), ethene (ETH, 75.0%), internal olefin carbon bond (IOLE, 70.4%), Ethane (ETHA, 55.7%), Xylene (XYL, 45.2%) and terminal olefin carbon bond (OLE, 42.2%) when using MOBILE6.2-Mexico, whereas this dominance is only maintained for BENZENE (57.6%) and ETH (52.4%) when applying MOVES-Mexico.

$PM_{2.5}$ emissions are mainly dominated by miscellaneous component of $PM_{2.5}$ (PMFINE, 61.2%) due to the large contributions of resuspension and construction activities (29% and 27%, respectively). Emissions from primary organic carbon (POC) and primary elemental carbon (PEC) account for 23.2% and 10.6% of the $PM_{2.5}$ emissions. In both cases, mobile sources, particularly Diesel Tractor Units, are the main sector associated to these emissions (24% and 41%, respectively). PSO4 and PNO3 are the lowest contributors with 3.9% and 1.1%.

In the case of NO_x, 92% of total emissions are related to NO and 6.6% and 0.7% to NO_2 and HONO, respectively (not shown).

Table 4
Contributions of area, mobile and point sources to total CB05-VOC and CB05-$PM_{2.5}$ species when using MOBILE6.2-Mexico (MBL6) and MOVES-Mexico (MVS) in the MCMA for the case study.

CB05-VOC specie	Area	Mobile (MBL6)	Point	Area	Mobile (MVS)	Point
ALD2	58.7%	41.1%	0.2%	79.3%	20.4%	0.3%
ALDX	89.4%	4.1%	6.4%	91.0%	2.5%	6.6%
BENZENE	11.7%	79.8%	8.5%	24.5%	57.6%	17.8%
ETH	8.5%	75.0%	16.5%	13.9%	52.4%	33.7%
ETHA	41.2%	55.7%	3.2%	64.7%	30.3%	5.0%
ETOH	84.7%	0.0%	15.3%	84.7%	0.0%	15.3%
FORM	53.1%	41.8%	5.1%	71.1%	21.9%	7.0%
IOLE	29.5%	70.4%	0.1%	52.6%	47.1%	0.2%
ISOP	97.4%	2.5%	0.1%	98.9%	0.9%	0.2%
MEOH	99.7%	0.0%	0.3%	99.7%	0.0%	0.3%
OLE	38.5%	42.2%	19.3%	52.6%	20.3%	27.1%
PAR	50.5%	47.0%	2.5%	73.1%	23.3%	3.6%
SESQ	100.0%	0.0%	0.0%	100.0%	0.0%	0.0%
TERP	65.1%	0.0%	34.9%	65.1%	0.0%	34.9%
TOL	62.9%	31.4%	5.7%	79.4%	13.4%	7.1%
UNR	57.8%	40.0%	2.2%	78.4%	18.6%	3.0%
XYL	43.6%	45.2%	11.3%	62.3%	21.6%	16.1%
CB05-$PM_{2.5}$ specie	Area	Mobile (MBL6)	Point	Area	Mobile (MVS)	Point
PNO3	8.6%	72.3%	19.1%	6.5%	79.3%	14.3%
POC	30.2%	51.8%	17.9%	28.2%	55.0%	16.8%
PSO4	20.2%	37.8%	42.1%	18.5%	43.0%	38.5%
PEC	20.5%	73.6%	6.0%	19.1%	75.4%	5.6%
PMFINE	38.9%	36.5%	24.7%	37.6%	38.6%	23.8%

3.2. Comparison of modelled concentrations to observations

Fig. 6 shows the time series of the average modelled and observed hourly CO, NO_2, O_3 and $PM_{2.5}$ concentrations during February 14–28 at all the RAMA stations. The red line denotes the air quality simulation run with the MOBILE6.2-Mexico traffic emissions, while the blue line indicates the concentrations modelled when using the MOVES-Mexico emissions. On the other hand, Fig. 7 shows scatter plots of the CO, NO_2 and O_3 pair measurement-model on an hourly basis at the urban traffic station of Merced (MER) and the suburban station of Villa de las Flores (VIF). Red and blue dots symbolise the results obtained using MOBILE6.2-Mexico and MOVES-Mexico emissions, respectively. $PM_{2.5}$ is not included due to a lack of data. In all cases, modelled outputs are presented without any correction factor or post-processing.

Statistical parameters on an hourly basis (i.e. mean bias, MB; root mean square error, RMSE; correlation coefficient, r; Index of Agreement, IOA) are presented for each pollutant according to the four categories of stations (Table 5). A description of the model evaluation statistics used is reported in Carslaw (2015).

Evaluation of PM_{10} is not included in the present work since the current version of HERMES-Mex does not account for local fugitive dust emissions caused by wind erosion (i.e. windblown emissions), which have been reported as a major contributor to PM_{10} concentrations in the MCMA (e.g. Querol et al., 2008; Díaz-Nigenda et al., 2010).

Regarding CO, the model follows well the variation of concentrations ($r = 0.69$), especially in urban traffic stations where CO emissions are produced locally ($r = 0.75$). The use of MOVES-Mexico emissions significantly reduces the overestimation of daytime peaks modelled with MOBILE6.2-Mexico, the MB in urban traffic stations being decreased from 0.6 ppm to −0.1 ppm and the IOA being increased from 0.25 to 0.67. This reduction can be clearly observed in the MER station (Fig. 7), in which the RMSE is reduced from 1.1 ppm to 0.4 ppm (not shown). On the other hand, the underestimation observed in suburban

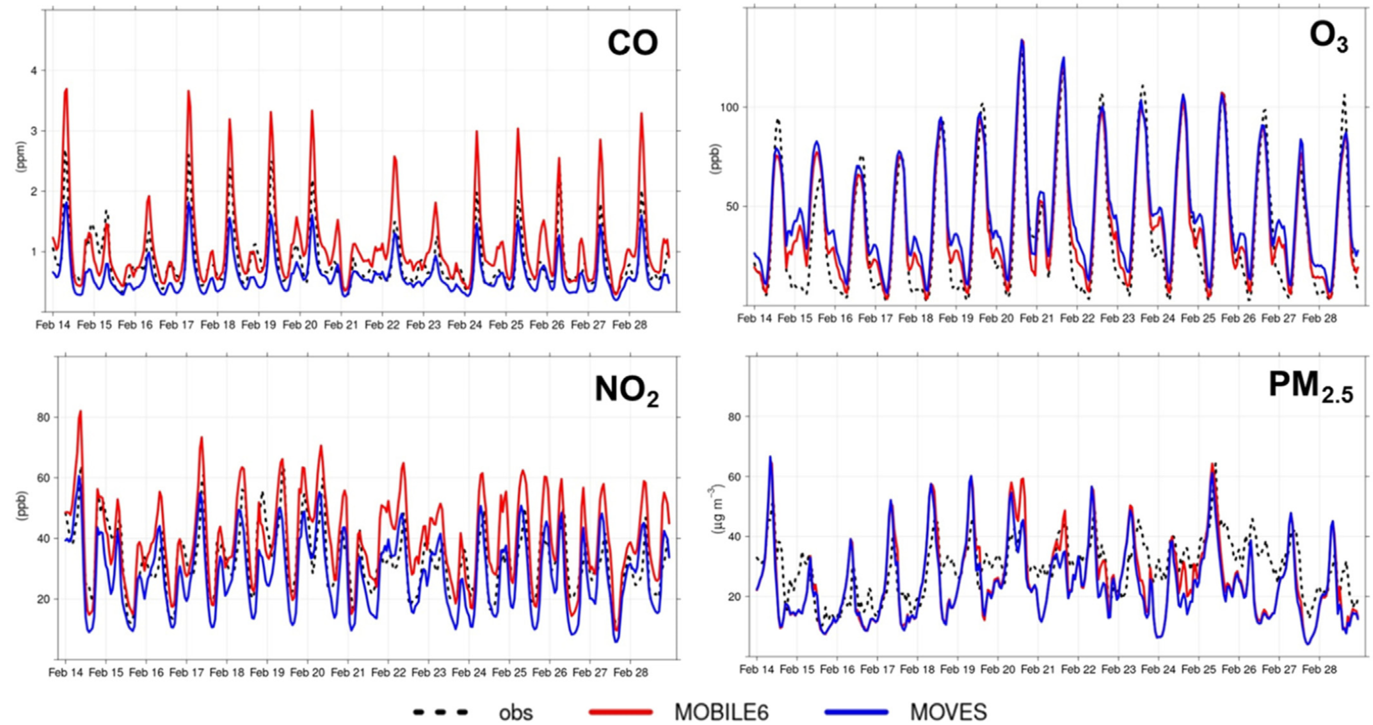

Fig. 6. Comparison of measured (black dashed line) and modelled (MOBILE6.2-Mexico, red line; MOVES-Mexico, blue line) diurnal profiles of surface hourly CO, NO_2, O_3 and $PM_{2.5}$ concentrations averaged over all monitoring sites. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

stations such as VIF is increased when using MOVES-Mexico due to the reduction of CO traffic emissions, the MB varying from −0.1 ppm to −0.4 ppm. It is also important to note that the model is capable of reproducing the weekend effect, the decrease in concentration from workdays to Sundays (16th and 23rd February) being ~30%.

NO_2 concentrations are correctly reproduced by the model, with a first peak reached during the morning due to the increase in traffic emissions during rush- hour and a secondary maximum seen close to midnight. In between these two peaks, concentrations first decrease and afterwards steadily increase again as a consequence of the evolution of the boundary layer growth (Pérez-Vidal and Raga, 1998). As in the case of CO, NO_2 MB and RMSE are highly reduced in urban traffic stations when replacing MOBILE6.2-Mexico emissions by MOVES-Mexico emissions (MB from 18.2 ppb to 5.5 ppb and RMSE from 25.5 ppb to 18.0 ppb). Fig. 7 shows this improvement in the MER station, where modelled concentrations are reduced by up to 50 ppb when using the MOVES-Mexico emissions. Errors in modelled NO_2 are somewhat larger in suburban stations than in urban stations, as demonstrated by the lower correlation coefficient and IOA. The weekend effect is also observed for this pollutant, indicating a good characterization of the temporal profiles applied to the traffic emission sources. It is important to note that part of the inconsistencies between modelled and observed concentrations could also come from the NO_2/NO_x ratios assumed for mobile sources (Section 2.6), which are based on European sources due to a lack of local references.

The model properly reproduces the temporal variations of O_3 concentrations ($r = 0.83$) and predicts most of the peak O_3 values observed, especially the ones recorded during the activation of the pre-contingency alert (i.e. February 20). The correlation coefficient and IOA values reach 0.9 and 0.8 in urban traffic stations (using both MOBILE6.2-Mexico and MOVES-Mexico), indicating that modelled concentrations agree well with measurements. The model shows an improvement of the MB and RMSE in urban traffic stations when applying MOVES-Mexico (MB = −0.5 ppb and RMSE = 17.4 ppb). This is largely due to the enhancement of the reproduction of high concentrations. As observed in the MER station (Fig. 7), modelled concentrations above 80 ppb are generally increased when applying MOVES-Mexico emissions. On the other hand, night-time values are generally overestimated, especially in suburban/rural areas and when applying the MOVES-Mexico emissions due to the reduction of fresh NO emissions. The highest uncertainties are observed in suburban stations, in which MB and RMSE reach values of 12.7 ppb and 24.0 ppb when using MOVES-Mexico. The overestimation of O_3 concentrations during the night of 14th February (which was not observed in the urban traffic stations) was rather large due to an overestimation of the modelled wind speed that transported an air mass with high O_3 concentrations outside of Mexico City (Supplementary material, Fig. 8).

Regarding $PM_{2.5}$, the model generally tracks the temporal variations of concentrations reasonably well, but frequently underestimates during the afternoon and nighttime when background levels are significantly influenced by anthropogenic secondary organic aerosols (SOA) (Vega et al., 2010). As pointed out by several works (e.g. Dzepina et al., 2009; Li et al., 2011) the traditional approach to predict SOA formation from VOC precursors (mainly aromatics) and oxidants (i.e. O_3 and OH^-), such as the one implemented in CMAQ (Carlton et al., 2010), fails to produce enough SOA to match the observations in the MCMA. The difference between using MOBILE6.2-Mexico and MOVES-Mexico is minimum, which is in line with the small differences observed in terms of primary emissions (Table 3). The MB slightly increase when using MOVES-Mexico instead of MOBILE6.2-Mexico (−5.7 versus −4.1 $\mu g \cdot m^{-3}$), which may be related to the reduction of NO_2 and subsequently nitrate formation.

Stations located in the urban area of the MCMA, in which emissions are produced locally, generally exhibit a better performance than those stations located in the suburbs of the MCMA. Two likely reasons for this are, on the one hand, failure of the pollutant dispersion processes due to an overestimation (~2.5 $m \cdot s^{-1}$) of the afternoon wind speed peak modelled by WRF (e.g. Zhang and Dubey, 2009; Li et al., 2011 and Fig. 8 of the Supplementary material) and, on the other, the non-consideration of emission sources that have a greater impact in the peripheral area of the MCMA than in the urban core region, such as biomass or trash burning (Hodzic et al., 2012). In the case of O_3, the difference in

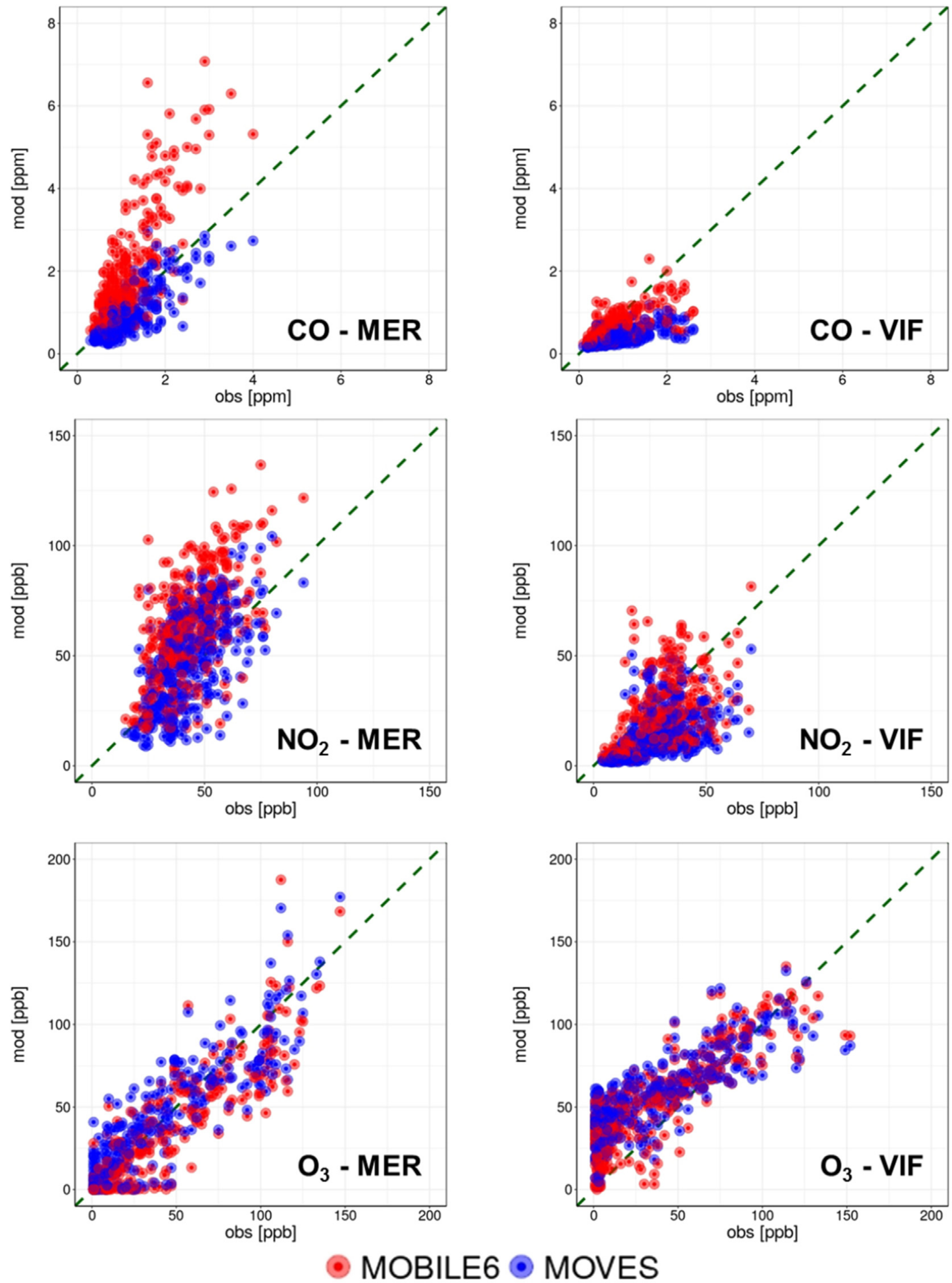

Fig. 7. Scatter plots of the CO, NO_2 and O_3 pair measurement model on an hourly basis at the urban traffic station of Merced (MER) and the suburban station of Villa de las Flores (VIF). Red and blue dots symbolise the results obtained using MOBILE6.2-Mexico and MOVES-Mexico emissions, respectively. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

the model behavior between rural/suburban and urban environments when it comes to reproducing the night-time destruction of O_3 is an already know characteristic of CMAQ, which performs a poor reproduction of O_3 titration in NO_2-limited regimes (Mao et al., 2010; Baldasano et al., 2011).

3.3. Comparison of modelled and observed CB05-VOC/CO ratios

Table 6 presents a comparison of predicted and observed CB05-VOC species to CO ratios at the MER and Pedregal (PED) stations. The comparison is limited to the morning period (i.e. 06:00 h–09:00 h), when concentrations are strongly related to anthropogenic emissions and less affected by the photochemistry (Velasco et al., 2007). Since the measured data is restricted to 22 Non-Methane Hydrocarbons (NMHCs) (i.e. 9 alkanes, 7 olefins, 6 aromatics) only 9 CB05-VOC species are considered in the comparison, namely ETHA, ETH, PAR, OLE, IOLE, isoprene (ISOP), BENZENE, TOL and XYL. A conversion of the measured VOC specie concentrations to the CB05 species was performed following Carter (2016) to allow a direct comparison.

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Table 5
Statistics obtained with MOBILE6.2-Mexico (MBL6) and MOVES-Mexico (MVS) on an hourly basis for the pollutants (p) NO_2, CO, O_3 and $PM_{2.5}$ for the study period (14–28 February 2014). Statistics are calculated according to four categories (c): urban stations with dominant traffic emission sources (URB-TRF), other urban stations (URB), suburban stations (SUB) and rural stations (RUR). The "n" column indicates the number of pair measurement-model used to compute the statistics. The calculated statistics are mean bias (MB), root mean square error (RMSE), correlation coefficient (r) and index of agreement (IOA).

			MB (*)		RMSE (*)		r		IOA	
p	c	n	MBL6	MVS	MBL6	MVS	MBL6	MVS	MBL6	MVS
NO_2	all data	8237	6.6	−4.5	20.6	17.3	0.65	0.63	0.35	0.45
	URB-TRF	1718	18.2	5.5	25.5	18.0	0.63	0.58	0.13	0.40
	URB	4089	6.0	−5.7	19.8	17.0	0.62	0.61	0.37	0.45
	SUB	1764	−2.3	−11.0	18.1	18.4	0.55	0.52	0.37	0.34
	RUR	666	4.4	−5.6	16.8	13.6	0.67	0.65	0.38	0.48
CO	all data	6119	0.3	−0.2	0.8	0.5	0.67	0.69	0.41	0.60
	URB-TRF	1716	0.6	−0.1	1.1	0.5	0.70	0.75	0.25	0.67
	URB	2354	0.2	−0.2	0.7	0.5	0.64	0.64	0.44	0.58
	SUB	1405	−0.1	−0.4	0.5	0.5	0.66	0.67	0.54	0.45
	RUR	644	0.3	0.0	0.4	0.2	0.73	0.71	0.32	0.66
O_3	all data	7143	3.7	10.1	20.1	22.1	0.83	0.83	0.74	0.71
	URB-TRF	1339	−6.3	−0.5	18.1	17.4	0.89	0.89	0.80	0.80
	URB	3735	5.7	12.3	20.2	22.7	0.84	0.83	0.74	0.69
	SUB	1405	6.2	12.7	21.5	24.0	0.82	0.83	0.72	0.68
	RUR	664	7.6	13.8	20.0	22.3	0.80	0.81	0.68	0.64
$PM_{2.5}$	all data	3803	−4.1	−5.7	17.7	17.6	0.41	0.38	0.39	0.38
	URB-TRF	1389	0.0	−1.5	16.8	16.7	0.45	0.41	0.38	0.39
	URB	2414	−6.6	−8.2	18.4	18.5	0.41	0.39	0.39	0.38
	SUB	1751	−15.0	−15.0	35.0	35.0	0.47	0.47	0.64	0.64
	RUR	0	–	–	–	–	–	–	–	–

* ppm for CO, ppb for NO_2 and O_3 and $\mu g \cdot m^{-3}$ for $PM_{2.5}$.

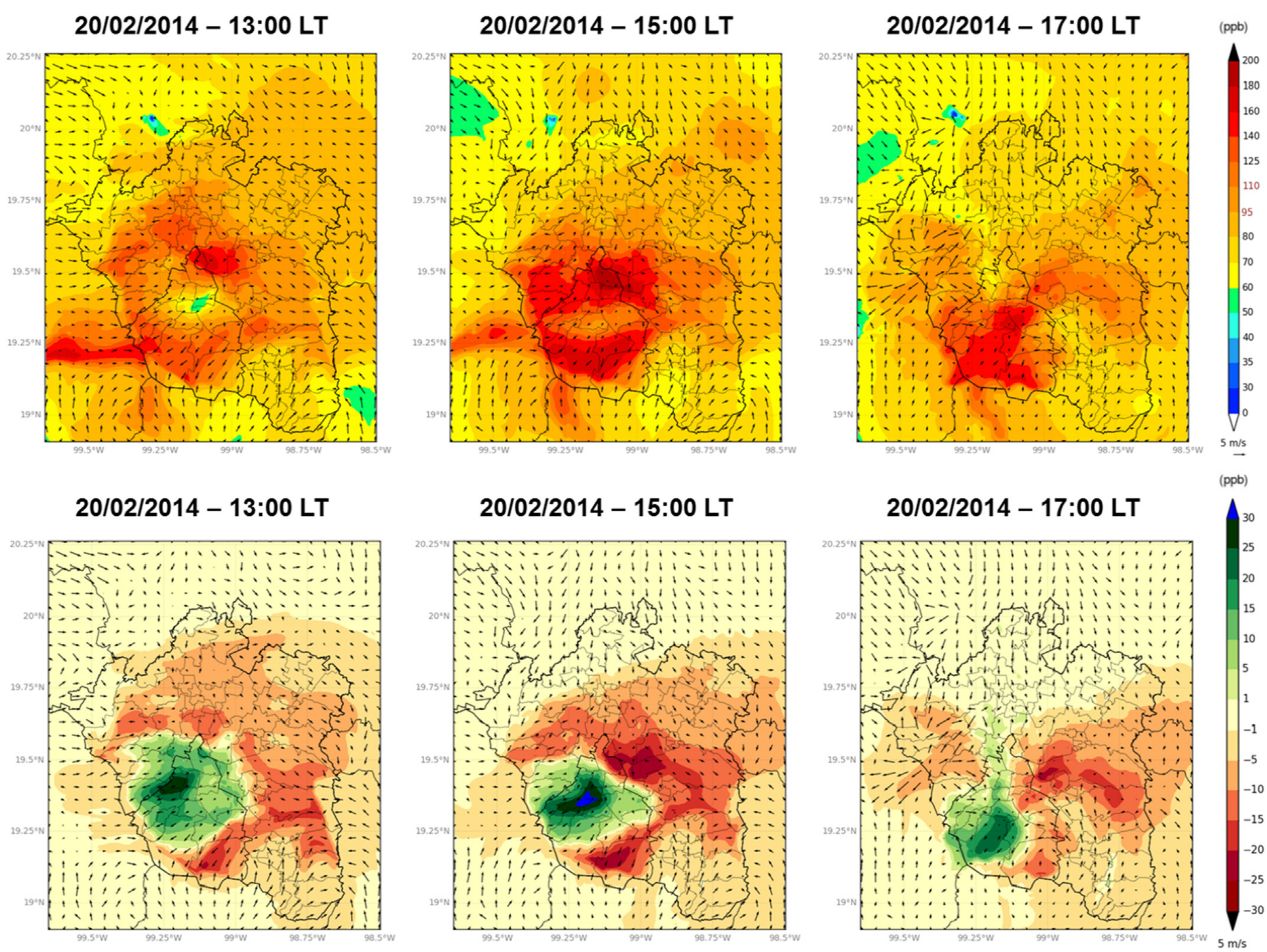

Fig. 8. Spatial distribution of modelled O_3 concentrations (ppb) at 13:00, 15:00 and 17:00 LT over the MCMA throughout the 20th February 2014 (top) and the difference when using MOVES-Mexico on-road emissions (MOVES-Mexico – MOBILE6.2-Mexico) (bottom). Black arrows simulate surface winds.

Table 6
Comparison of observed (obs) and modelled (MOBILE6.2-Mex, MBL6; MOVES-MEX, MVS) morning (06:00–09:00) CB05-VOCs/CO ratios at La Merced (MER) and Pedregal (PED) stations. Values in italics indicate hourly mean CO concentration observed and modelled at each station.

CB05-VOC (ppbV)	MER			PED		
	CO (ppmV)			CO (ppmV)		
	obs *1.2*	MBL6 *3.8*	MVS *1.7*	obs *0.7*	MBL6 *1.7*	MVS *0.9*
ETHA	9.1	3.3	4.8	11.8	2.8	3.6
ETH	13.4	7.9	9.5	15.5	7.0	7.6
PAR	326.0	254.5	354.5	324.2	240.0	307.6
OLE	11.9	4.7	6.8	12.2	3.6	4.4
IOLE	2.0	0.6	0.6	2.9	0.5	0.4
ISOP	1.0	0.3	0.3	0.8	0.3	0.3
BENZENE	1.2	1.6	1.6	1.3	1.5	1.5
TOL	7.8	9.2	13.9	7.0	8.1	10.7
XYL	2.5	5.5	6.4	2.0	4.8	4.9

Generally speaking, ratios computed using MOVES-Mexico emissions are more in line with observations than the ones modelled when using MOBILE6.2-Mexico, except for some aromatics (i.e. TOL and XYL). As seen in the previous section, the system presents a better performance in the urban core area (MER station) than in the surroundings of the city center (PED station). Note that the overestimation of CO concentrations when using MOBILE6.2-Mexico is significantly reduced when using MOVES-Mexico in the two stations (Table 6, values in italics).

The modelled BENZENE/CO ratios remain the same and show a good agreement with observations in all cases. Nevertheless, when using MOBILE6.2-Mexico the overestimation of CO concentrations is compensated by an overestimation of BENZENE concentrations. Considering that BENZENE is mainly emitted by traffic sources (Table 4), this result suggests that the estimated on-road mobile source VOC emissions may be more accurate when using MOVES-Mexico. This conclusion can be also extrapolated from the modelled ETH/CO ratios. Some other CB05-VOC species that are overestimated when using MOBILE6.2-Mexico (i.e. IOLE, OLE) become slightly underestimated when using MOVES-Mexico mobile emissions. Suggesting that there may be missing area and point sources in the inventory for these species. Results show that TOL/CO and XYL/CO ratios remain overestimated despite reducing mobile VOC emissions, indicating that emissions from the main contributors to these species (i.e. use of solvents in architectural paints, industrial and other coating applications) may be revised. The systematic underestimation of the observed ISOP/CO ratios may be related to an underestimation of biogenic isoprene due to either an incorrect characterization of local emission factors or the underestimation of maximum temperature values modelled by WRF (Supplementary material).

As mentioned before, there is a 2 year differences between observed and modelled data and parameters such as different meteorological conditions may be influencing the results presented.

3.4. Ozone formation

Despite reducing the estimation of modelled O_3 precursor emissions when using MOVES-Mexico instead of MOBILE6.2-Mexico (Table 3) the changes of episode peak O_3 concentrations are very low (between 13:00 and 17:00 LT). The reduction of total NO_x (−37%) is larger than the one observed for toluene (−21%), which has been reported as the main VOC contributor to O_3 formation in the MCMA (Garzón et al., 2015). This difference in the decrease of precursor emissions leads to a 3.2% increase (2.5 ppb) in the O_3 peak averages in the mean of all stations, the variations observed in the urban traffic monitoring stations being largest (6.6%, 5.2 ppb) (Table 7). As pointed out by Zavala et al., (2009b), O_3 concentrations in Mexico City are more sensitive to perturbations of all emissions other than mobile source alone (e.g. the use of solvents and distribution of LPG). When analyzing individual days it is observed that differences of O_3 peak averages present a wide range of variation, with increases of 0.4% (20th February) to 6.7% (27th February). This fact indicates that changes in O_3 peak concentrations are highly dependent on the meteorological conditions of each day. To confirm this hypothesis, both days are analysed in more detail in the following paragraphs.

Figs. 8 and 9 shows the spatial distribution of O_3 concentrations for the 20th and 27th February at 13:00, 15:00 and 17:00 LT when using MOBILE6.2-Mexico (top), as well as the difference when changing to MOVES-Mexico emissions (bottom). Since meteorological conditions play a key role in determining the dispersion or accumulation of pollutant concentrations in the MCMA (de Foy et al., 2005), the spatial distributions of modelled O_3 concentrations are presented along with the modelled wind fields.

Results show how the stagnant conditions in the MCMA on the 20th February are favourable for the build-up of high O_3 concentrations, which exceed the hourly limit value of 110 ppb over almost the entire urban region at 15:00 LT (Fig. 8, top). The O_3 episode can be partly associated with the "O_3-south" meteorological episode category defined by de Foy et al., (2008). In this case, a weak synoptic forcing associated with an anticyclone, as well as thermally driven circulations, lead to the formation of a convergence zone in the south of the MCMA and subsequent high O_3 peaks in this area. Nevertheless, and as a consequence of the weak northern winds, in the present case it is observed how a part of the modelled plume remains stagnant in the city centre (15:00 LT). At 17:00 LT the strong northwest winds start to push the plume to the south of the MCMA and the concentrations in the core urban area start to decrease.

On the other hand, on the 27th February it is observed how westerly and southerly winds transport the O_3 plume outside of the MCMA core urban area (northeast), the maximum concentrations reached in this area being lower than 70 ppb (Fig. 9, top). Considering the humid conditions observed on this day (Supplementary material, Fig. 7) and the rainfall that occurred in the northeast of the MCMA in the afternoon (Supplementary material, Fig. 4), this episode could be linked to the "O_3-north convective" condition also described by de Foy et al., (2008).

Compared to the base case (MOBILE6.2-Mexico emissions), O_3 concentrations increase in the urban core region along the east and west ridges bordering the city for both the 20th and 27th February while generally decreasing in mountain areas (up to ±30 ppb, Figs. 8 and 9, bottom). Nevertheless, results also show how for some specific suburban/rural regions, O_3 concentrations experience both increases and decreases, depending on the day of study. This fact indicates that changes in O_3 peak concentrations in the surrounding of MCMA urban area are highly sensitive to the meteorological episode, and particularly to the direction of the plume. This is in line with the results obtained by Song et al., (2010), which pointed out that the MCMA urban core area is VOC-limited, while the surrounding areas can be either a NOx- or VOC- limited regime, depending on the meteorological conditions.

4. Conclusions

This study presents the High-elective Resolution Modelling Emission System for Mexico (HERMES-Mex) model, an emission processing tool to support regional air quality modelling in the Mexico City Metropolitan Area (MCMA). A case study (14–28 February 2014, during which the O_3 environmental pre-contingency alert was activated) is performed to demonstrate the applicability of HERMES-Mex to provide Community Multi-scale Air Quality (CMAQ) model-ready high-resolution emissions. Air quality concentrations modelled by the system composed of the Weather Research and Forecasting model (WRF), HERMES-Mex and CMAQ are compared with measurements. Resulting traffic emissions from the Mobile Source Emission Factor Model for Mexico (MOBILE6.2-Mexico) and the MOtor Vehicle Emission Simulator for Mexico (MOVES-Mexico) are integrated in HERMES-Mex to assess and compare

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Table 7
Average and percentage changes of peak O_3 concentrations (13:00–17:00 h LT) in all stations and urban traffic stations due to change of traffic emissions from MOBILE6.2-Mexico (MBL6) to MOVES-Mexico (MVS) for the whole case study (14–28 February 2014) and the 20th and 27th February 2014.

	All Stations				Urban Traffic Stations			
	obs ppb	MBL6	MVS	diff %	obs ppb	MBL6	MVS	diff %
All episode (14–28 February) (13:00–17:00 LT)	87.8	85.6	88.3	3.2%	92.3	79.0	84.2	6.6%
20th February (13:00–17:00 LT)	112.7	121.2	121.7	0.4%	125.9	120.9	122.0	0.9%
27th February (13:00–17:00 LT)	52.9	61.2	65.3	6.7%	47.0	53.0	58.5	10.3%

their performance. The O_3 sensitivity to the change of traffic emissions is spatially analysed.

The main findings and conclusions of this work are as follows:

- When replacing MOBILE6.2-Mexico by MOVES-Mexico, total emission estimations in the MCMA are reduced for NO_x (−37%), CO (−52%) and VOCs (−26%), while slightly increased for PM_{10} (+8%) and $PM_{2.5}$ (+6%). Gasoline taxis and passenger cars are the categories that contribute the most to the reduction of NO_x, CO and VOC on-road emissions (between −39% and −90%), while their large increases in PM_{10} and $PM_{2.5}$ (up to +180%) are compensated by the decrease of Diesel tractor unit emissions (−40%). The most significant emission decrease occurs in downtown Mexico City due to the higher presence of traffic activity.
- The WRF/HERMES-Mex/CMAQ system properly reproduces the concentration levels and patterns of CO, NO_2, and O_3 in the MCMA. The performance of the system tends to be better in those stations located in the core urban area than in those located in the suburbs of the MCMA. Two likely reasons for this are, on the one hand, a bad characterization of the pollutant dispersion processes modelled by WRF and, on the other, the non-consideration of emission sources that have a greater impact in the peripheral area of the MCMA than in the urban core region, such as biomass or trash burning. In the case of $PM_{2.5}$ an underestimation of afternoon levels indicates potential uncertainties with the processes related to the formation of anthropogenic secondary organic aerosols (SOA).
- The system's performance clearly improves in urban stations with a strong influence of traffic sources when changing from MOBILE6.2-

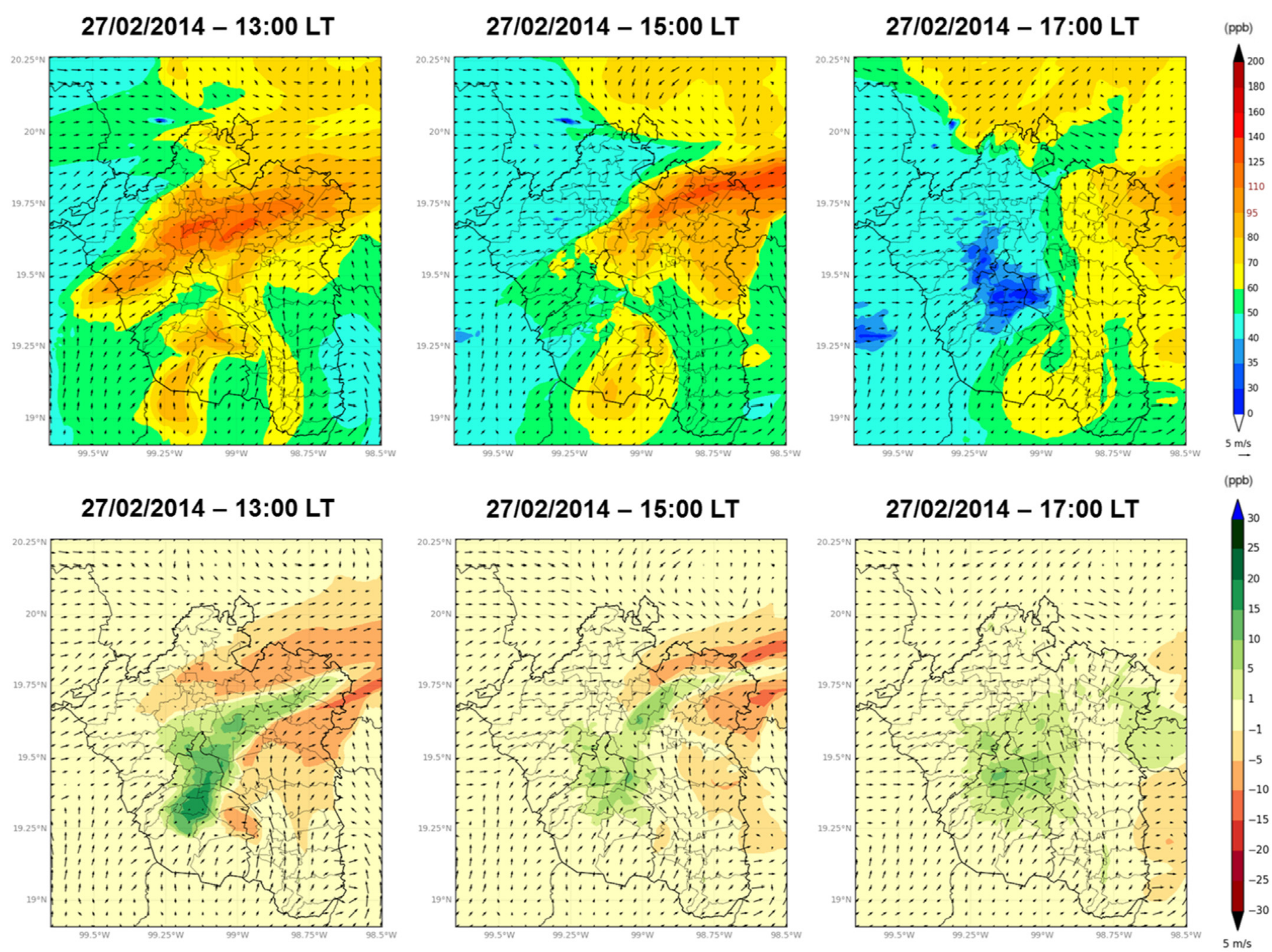

Fig. 9. Spatial distribution of modelled O_3 concentrations (ppb) at 13:00, 15:00 and 17:00 LT over the MCMA throughout the 27th February 2014 (top) and the difference when using MOVES-Mexico on-road emissions (MOVES-Mexico – MOBILE6.2-Mexico) (bottom). Black arrows simulate surface winds.

Mexico to MOVES-Mexico traffic emissions. The MB and RMSE observed for NO_2 are significantly reduced (MB from 18.2 ppb to 5.5 ppb and RMSE from 25.5 ppb to 18.0 ppb), while in the case of CO the correlation coefficient and IOA are also enhanced (r from 0.70 to 0.75 and IOA from 0.25 to 0.67). These results reinforce the conclusions reached by previous works that stated that MOBILE6.2-Mexico NO_x and CO emission factors were overestimated. In the case of O_3, MB is also reduced (from −6.3 ppb to −0.5 ppb) while the correlation does not change (0.89). For $PM_{2.5}$ MB is slightly increased when applying MOVES-Mexico ($-4.1\ \mu g \cdot m^{-3}$ to $-5.7\ \mu g \cdot m^{-3}$), probably due to the reduction of NO_2 and subsequently nitrate formation.
- VOC species that are mainly associated to mobile sources (i.e. benzene, ethene) are more in line with observations when using MOVES-Mexico instead of MOBILE6.2-Mexico. Remaining inconsistencies for olefins and some aromatics (e.g. toluene, xylene) may indicate that area source emissions (i.e. use of solvents in architectural paints, industrial and other coating applications) should be revised.
- Average peak O_3 concentrations are increased by 6.6% (urban traffic stations), when using MOVES-Mexico instead of MOBILE6.2-Mexico emissions, showing a larger consistency with observations. Peak O_3 concentrations are increased in the urban core region while decreased in mountain/rural areas (by up to ±30 ppb). As suggested in previous works, the MCMA urban core area remains VOC-limited during the whole episode, while the surrounding areas can be either a NO_x- or VOC- limited regime, depending on the meteorological conditions.
- These results suggest that in order to reduce O_3 concentrations in Mexico City, emission control policies of mobile sources should be simultaneously combined with reductions of those activities related to the use of solvents and the distribution of LPG. Nevertheless, and in order to better assess this hypothesis, a longer period of study should be simulated using HERMES-Mex as a tool for emission scenario analysis and evaluation of air quality management strategies.

The results obtained suggest that HERMES-Mex can be used to provide model-ready emissions for air quality modelling in the MCMA. The conclusions shown in this work are derived from the study of an air pollution episode in 2014 (worst-case). Although it cannot be considered a large period, it is an episode with significant air pollution. Future studies should include a longer (i.e. annual) evaluation period of the WRF/HERMES-Mex/CMAQ system in order to reinforce the conclusions presented and test it over other typical meteorological conditions of the MCMA (i.e. rainy and warm-dry season). This future evaluation work should include the analysis of $PM_{2.5}$ components and a more complete VOC sample to provide additional hints on the uncertainties related to the system's performance.

Regarding the improvement of the HERMES-mex model, future works will be related to the inclusion of other potential pollutant activities not currently considered (i.e. biomass and trash burning, and dust emissions related to soil erosion processes), as well as the implementation of a process to allow plume rise calculation and the performance of sensitivity tests related to the speciation and vertical injection of primary emissions. The HERMES-Mex model will constitute the emission core of the Air Quality Forecast System for the City of Mexico (AQFS-MexDF), being currently under development by the Barcelona Supercomputing Center (BSC) and the Mexico City's Secretariat of the Environment (SEDEMA).

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Appendix A. Supplementary data

Supplementary data to this article can be found online at http://dx.doi.org/10.1016/j.scitotenv.2017.01.135.

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