Vehicle-Induced Fugitive Particulate Matter Emissions in a City of Arid Desert Climate

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Abstract

This study investigates and proposes emission factors (EFs) and models for vehicle-induced exhaust (VEX) and fugitive (VfPM) particulate matter emissions representative of areas with arid climates. Particle number (PNC) and mass (PMC) concentrations and their integrated samples were collected for a period of three months for both PM₁₀ and PM_{2.5} next to a trafficked

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road in the city of Doha, Qatar. Using Positive Matrix Factorization (PMF) on the elemental data of the samples, six distinct PM sources were identified: traffic exhaust, dust resuspension, fresh and aged sea salt, secondary aerosols, and fuel oil/shipping. Dispersion modelling and regression analysis were combined to derive EFs (linear analysis) and models (non-linear analysis) for the total traffic fleet (heavy and light duty). The estimated EFs were between 620 to 730 mg VKT⁻¹ (VKT; Vehicle Kilometer Travelled) (adj. R² ~ 0.84) and 1080 to 1410 mg VKT⁻¹ (adj. R² ~ 0.70) for VEX and VfPM, respectively. The integration of field measurements, chemical characterization, and dispersion modelling presented herein, is one of the first similar studies conducted in the wider region, identifies the importance of fugitive PM (fPM), and marks the need for further studies to improve emissions modelling of VfPM in arid desert climates.

Keywords: Vehicle-induced particulate matter; fugitive emissions; source apportionment; emission factors; arid desert climates

1. Introduction

Road traffic is an indisputable source of particulate matter (PM) pollution in urban areas (Pant and Harrison, 2013). Exposure to vehicular PM can cause acute respiratory and cardiopulmonary conditions, especially to sensitive groups such as children and elders (Brauer, 2002; Fan et al., 2009). Therefore, exhaust particles that come out of a vehicle's tailpipe have been extensively studied and widely regulated in many European and North American countries (Thorpe and Harrison, 2008). Over the years, implementation of emissions control strategies has led to successful reduction in exhaust PM. In contrast, little has been done to address the impact of the non-exhaust (i.e. fugitive) sources (Shirmohammadi et al., 2017).

Vehicle-induced fugitive Particulate Matter (VfPM) is generated from the abrasion of brake, tire and road components as well as resuspension of dust by the passing vehicles (Kumar et al., 2013). The latter being of greater importance in areas and urban environments with desert arid climate (Tsiouri et al., 2015). Studies in North Carolina, USA suggested that VfPM is the main source of coarse particles (PM_{10} ; $\leq 10 \mu m$ in aerodynamic diameter) in urban roads (Abu-Allaban et al., 2003). Measurements collected from two different types of roads in Switzerland found that brake wear and road dust contributed 59% of the total PM_{10} , compared to 41% from exhaust sources (Bukowiecki et al., 2010). Furthermore, abrasion of brake and tire components can produce enormous amounts of sub-micron sized particles ($<300 \text{ nm}$ in diameter), which pose a higher risk on human health (Kumar et al., 2013). Despite their significance in urban roads, the current knowledge on VfPM and their behavior is far from comprehensive (Pant and Harrison, 2013).

Various techniques have been developed to measure and characterize VfPM. The conventional techniques include quantifying VfPM under real-time conditions through

simultaneous measurements at a road-side and a background location ([Harrison et al., 2012](#); [Krecl et al., 2018](#)), measuring concentrations at the ends of a road tunnel with known boundary conditions ([Lawrence et al., 2016](#); [Riccio et al., 2016](#)) or collecting direct measurements using mobile vehicles ([Hussein et al., 2008](#); [Kwak et al., 2014](#)). In reality, estimating individual contributions from non-exhaust (VfPM) traffic sources is difficult because of the interactions between the different sources (e.g. differentiating between the wear directly emitted by the road surface and the pre-deposited material on the road) ([Thorpe and Harrison, 2008](#)). Chemical characterization and source apportionment methods are often combined to determine source-specific contributions through the fingerprints of their tracer elements ([Amato et al., 2016a](#)). For instance, [Bukowiecki et al. \(2010\)](#) carried out an elemental size-segregated measuring campaign at two different roads in Switzerland, where the US EPA Positive Matrix Factorization (PMF) model was employed to identify source contributions and estimate PM₁₀ emission factors (EFs) for brake wear and resuspension using pre-estimated NO_x EFs for Switzerland ([INFRAS, 2007](#)). Other studies aimed to examine a particular mechanism under a controlled environment, such as the use of laboratory scale models to simulate road-tire interactions ([Grigoratos et al., 2018](#)). Laboratory set ups are advantageous in maximizing the collection of particles, but they tend to underestimate emissions, probably due to the elimination of real world dilution effects ([Aatmeeyata et al., 2009](#); [Dahl et al., 2006](#)).

The amount and characteristics of emitted VfPM are highly dependent on meteorology, surface characteristics, and driving conditions. For example, the use of studded tires during the winter season in Nordic countries can dramatically increase the amount of VfPM ([Ketzel et al., 2007](#)). In the study of [Hussein et al. \(2008\)](#), mobile measurements were collected from different roads and pavements in Stockholm using a modified version of the TRAKER (Testing Re-

entrained Aerosol Kinetic Emissions from Roads) test vehicle (described in Etyemezian et al., 2003). The results showed that studded tires increase road dust emissions between 2.0 to 6.4 times compared to friction tires and 4.4 to 17 times compared to summer tires. In contrast, a recent study by Lawrence et al. (2016) estimated non-exhaust PM_{10} EFs from a busy tunnel in London to be just slightly higher (i.e. 16.7-19.3 mg VKT⁻¹ – vehicle kilometer travelled) compared to exhaust emissions (i.e. 11.1-12.8 mg VKT⁻¹). In a similar way, resuspension of road dust during wet conditions (e.g. rain or snowfall) is low, compared to dry periods when evaporation rates increase with temperature (Omstedt et al., 2005).

Surely, there is a large number of specific EFs available in the literature to calculate VfPM emissions based on road type, vehicle class, and driving conditions (some are summarized in Table 1), mostly developed for the typical fleets and climate conditions of Europe and North America. It is obvious, however, that these are so specialized that they might be restricted to specific regions, and hence, non-applicable in areas where fugitive Particulate Matter (fPM) tend to be a bigger challenge like arid or semi-arid desert climates (Hassan et al., 2016). For this reason, the overall aim is to generate EFs for VfPM appropriate for urban environments in desert arid climates.

In this work, EFs and models for such an urban environment (i.e. city of Doha, Qatar) and representative of the wider Middle East area, have been derived by utilizing field measurements taken nearby major roads, source apportionment to estimate source contributions, dispersion modeling and regression analysis. The results presented in this work aim to contribute to a more comprehensive emission inventory for fPM in desert arid and semi-arid areas.

101 **Table 1** Literature Emission Factors (EFs) for Traffic PM [mg VKT⁻¹]

Location	Road Characteristics	Methodology	Exhaust	Non-exhaust	Reference
Nevada and North Carolina, USA	Different roads with high speed, low speed and exits	Roadside continuous measurements were used to derive total fleet emission factors, and integrated PM samples were used to attribute emission factors to exhaust, resuspended road dust and brake wear emissions using Chemical Mass Balance (CMB) modeling.	PM _{2.5, LDV} : 7-52 PM _{2.5, HDV} : 57-480 PM _{10, LDV} : 7-65 PM _{10, HDV} : 57-570	Brake Wear: PM _{2.5, LDV} : 0-4 PM _{2.5, HDV} : 0-13 PM _{10, LDV} : 0-79 PM _{10, HDV} : 0-610 Resuspension: PM _{2.5, LDV} : 2-26 PM _{2.5, HDV} : 17-300 PM _{10, LDV} : 41-780 PM _{10, HDV} : 230-7800	(Abu-Allaban et al., 2003)
Treasure Valley (TV), Idaho, USA	Arteria, Collector, Interstate, Local/Residential	Vertical measurements of PM were collected upwind and downwind a road. The measured concentrations were used to calculate emission factors based on wind speed and direction. PM were also measured in front and behind a moving vehicle's tire using Testing re-entrained aerosols kinetic emission from roads (TRAKER) method. Concentrations were linked to variables such as vehicle speed and downwind flux using regression analysis to estimated emission potential.	-	Resuspension: PM ₁₀ : 1700-8000	(Etyemezian, 2003)
Copenhagen, Denmark	Urban/Asphalt	Swedish Model; calculates emission factor for non-exhaust	PM: 28-50	PM ₁₀ : 46-198	(Ketzel et al., 2007)

Stockholm, Sweden Leipzig & Berlin, Germany Helsinki, Finland Klagenfurt, Austria		traffic emissions by accounting for the variability of dust load and surface moisture. (Omstedt et al., 2005)			
Copenhagen, Denmark	Urban (main road, traffic lights and slow traffic)	German Method; uses the procedure in INFRAS (2007) emission factor handbook to calculate exhaust emissions, and the procedure of Gehrig et al. (2004) to calculate non-exhaust traffic emissions using PM concentration difference upwind/downwind and NOx (dilution tracer).	PM: 19-32	PM _{10,fleet} : 29-118 PM _{10,LDV} : 10-90 PM _{10,HDV} : 200-800	
Copenhagen, Denmark	Urban/Asphalt	Danish Method; emission factors follow the structure of COPERT 4 model (Gkatzoflias et al., 2006), and depend on vehicle class (e.g. car/van/truck)	PM: 50	PM _{10,fleet} : 59 PM _{10,LDV} : 50/70 PM _{10,HDV} : 230	
London, UK	Urban/Roadside, Street Canyon	Measurements. Estimating PM ₁₀ using NOx as exhaust tracer. Non-exhaust=coarse fraction PM _{2.5-10} . Resuspension=Coarse-Abrasion	-	Resuspension: PM _{10,LDV} : 0-48 PM _{10,HDV} : 139-432	(Thorpe et al., 2007)
Barcelona, Spain	Urban	PM vertical profiles were estimated using an array of samplers arranged in a vertical set up next to a road. The deposition and emission flux were estimated using the terminal concentration (calculated from the square fit of	-	Resuspension: PM ₁₀ : 12-47	(Amato et al., 2012)

all samplers concentrations).

2. Methodology

2.1 Derivation of the VfPM Emissions

Traffic induces PM emissions directly and indirectly. The former, referred to as vehicle exhaust (VEX) emissions, depend mainly on the vehicle numbers, type, technology and the driving conditions factors. The estimation of VEX emissions has been thoroughly studied with well-established literature, EFs and models like the European COPERT ([Gkatzoflias et al., 2007](#)) and the US MOVES ([USEPA, 2016](#)). The indirect traffic emissions (non-exhaust or VfPM) are the topic of this study. They are more complex and, in addition to the above factors, depend on the meteorology and road conditions (e.g. surface material, deposited dust). As noted in Section 1, one of the approaches to estimate the EFs is to combine source apportionment studies, based on field campaigns, with dispersion modelling tools, based on accurate traffic data. The source apportionment tool calculates the contribution of VEX emissions to the measured values of PM or gaseous pollutants by taking advantage of their very characteristic elemental profile. The dispersion modelling relates the vehicle numbers (traffic intensities) and the aforementioned factors with these contributions and provides the means to estimate the EFs. Including more factors into the calculations increases the accuracy of the EFs and decreases the uncertainty. The use of gaseous pollutants (e.g. using NO_x EFs to estimate dilution rate; ([Pant and Harrison, 2013](#))), when available, is preferred for PM.

On the other hand, barren lands, construction sites, and other natural sources of PM are expected to have a similar elemental profile to the road dust resuspension ([Amato et al., 2011](#)) - in other words to the VfPM. This prohibits direct estimation of the VfPM without further assumptions and calculations. One option to estimate the VfPM is to calculate the individual contributions of all non-traffic sources with a similar elemental profile using emissions and

dispersion modelling and subtract them from the total measured contribution in order to obtain the VfPM portion.

Usually, this total measured contribution is referred to as resuspension or crustal material in source apportionment studies. This option has been followed herein with the additional hypothesis that transboundary contributions to the airborne crustal material (i.e. resuspension) are strongly correlated with wind (speed and direction) or negligible under normal dust levels, i.e. excluding regional dust events. The latter is a critical hypothesis that will be investigated prior to the calculations, and was based on a recent study by Kontos et al. (2018) that focused on the natural dust emissions in the wider area of interest and a previous own study on the sensitivity of fPM emissions to land use and land cover data (Hassan et al., 2017; Hassan et al., 2016). A regression analysis with all the related factors (traffic intensities, vehicle class, and meteorology) will result in an appropriate EFs for VfPM with acceptable correlation to the measurements.

Figure 1 describes the above methodology and the interconnections between the employed methods, while each method is described in detail in the following sections.

Figure 1 Outline of the methodologies for the estimation of VEX EFs (orange pathway), VfPM EFs (green pathway), and the interconnections with the employed methods (blue blocks; in brackets the corresponding section number). Other results (white blocks w/ blue outline) and non-studied phenomena (greyed blocks) are also displayed.

2.2 Field Campaign

Measurements were carried out during a three-month field campaign in the city of Doha in Qatar. Qatar [11,571 km²] is a coastal state with ~ 2,750,000 inhabitants, where the majority live in or around its capital city, Doha. Doha represents 1.1% of Qatar's total area and accommodates ~ 40% of its total population. Qatar has witnessed a substantial growth in its transportation sector in the recent years. According to Qatar's ministry of development planning and statistics, the total number of registered vehicles (including motorcycles) have increased by 34% between the years 2011 and 2015 ([MDPS, 2015](#)).

The field campaign was conducted from April to June 2015 at a sampling site adjacent to a trafficked road. The monitoring site was set up just outside a high-rise office building, located at Al Rumaila West area at the city center of Doha (Figure 2), characterized by sparse high-rise buildings (around 3-7 floors). The building overlooks Al Istiqlal Road (~ 50 m), a busy urban road with an average fleet of 77,300 vehicles per day during weekdays and 54,400 vehicles per day on weekends with ~ 95% of the vehicles being light duty. The road has 6 lanes, 3 in each direction, and intersects with Mohammed Bin Thani road (NW) and Al Rayyan road (SW), which are two major roads of the city's road network.

Particle number (PNC) and mass (PMC) concentrations were measured using Grimm Enviro Check 365, an air quality monitoring station manufactured by Grimm Aerosol Technik GmbH & Co. KG, Germany ([Grimm, 2009](#)). The station uses light scattering technology to measure hourly averages of PNC [particles liter⁻¹] over a size range of 0.25 up to 32 µm in 31 size channels using a sampling rate of 1.2 liter min⁻¹. It produces PMC [µg m⁻³] using a

proprietary algorithm calibrated to the National Institute of Standards and Technology (NIST, USA) Arizona test dust. According to the vendor's certificate, measurements are equivalent to the European Standards EN12341 and EN14907. Meteorological parameters were recorded by the climate sensor (WS600-UMB by G. Lufft Mess- und Regeltechnik GmbH, Germany) attached to the top of the monitoring station. The accuracies of the meteorological measurements were $\pm 0.3 \text{ m s}^{-1}$ or $\pm 3\%$ (range 0-35 m s^{-1}) and $\pm 5\%$ ($>35 \text{ m s}^{-1}$) for wind speed, $\pm 0.2 \text{ }^{\circ}\text{C}$ for temperature (range -20-50 $^{\circ}\text{C}$), $\pm 2\%$ for relative humidity (range 0-100%), and $<3^{\circ}$ (RMSE $> 1.0 \text{ m s}^{-1}$; range 0-359.9 $^{\circ}$) for wind direction. The sampling inlet was at $\sim 1.7 \text{ m}$ height from the ground.

Integrated ambient samples of $\text{PM}_{2.5}$ and PM_{10} were collected daily (on 24h basis) using two separate MicroPNS air samplers manufactured by WB Engineering Germany GmbH following the EN12341:2014 standard (NSAI, 2014). Each sampler sucks in the particulate-laden ambient air stream with a continuously regulated flow through a size-selective inlet ($\sim$ at a height of 1.7 m) and retains the particulate matter on a 47 mm diameter filter. Tissue quartz filters (by PALL) were used to collect a total of 90 (24h) samples of $\text{PM}_{2.5}$ and PM_{10} . Prior and after each sampling, filters were conditioned for 24-48 hr at 20-23 $^{\circ}\text{C}$ and 30-35% RH (USEPA, 1987), and gravimetrically weighed using a weighing cell (Sartorius WZA26-CW by Sartorius Lab Instruments GmbH & Co, Germany) with 1 μg readability and $\leq \pm 1 \text{ } \mu\text{g}$ repeatability. After weighing, the filters were placed in petri-dishes and transported to and from the sampling site in sealed containers to avoid contamination. Samples were then chemically analyzed for carbon, ionic and elemental characterization. More details on the chemical characterization methods have been published in a past study of the authors (Saraga et al., 2017) and the elements/species employed herein are reported in Section 2.3.

Figure 2 A map of Doha's major roads network showing the sampling site and the adjacent roads

2.3 Source Apportionment

Source Apportionment is the identification of ambient air pollution sources and the quantification of their contribution to air pollution levels. Receptor modeling comprises a source apportionment approach, aiming to re-construct the contribution of emissions from different sources of atmospheric pollutants based on ambient measurement data (i.e. PM chemical composition) observed at monitoring sites. One of the most successfully applied receptor models (Belis et al., 2015) is Positive Matrix Factorization (PMF) (Paatero and Tapper, 1994). PMF introduces a weighting scheme considering errors of the data points, which are used as point-by-point weights. Adjustment of the corresponding error estimates also allows it to handle missing

and below detection limit (DL) data. Moreover, non-negative constraints are implemented in order to obtain more physically meaningful factors.

The latest PMF version available by US EPA (PMF v.5.0) has been used in the present study. PMF analysis background is described in details by Paatero (Paatero, 1997). In brief, the factor model can be written as:

$$X = GF + E \quad (1)$$

where X is the known $n \times m$ matrix of the m measured chemical species in n samples. G is an $n \times p$ matrix of the p sources contributions to the samples (time variations). F is a $p \times m$ matrix of source compositions (source profiles). Both G and F are factor matrices to be determined. E is defined as a residual matrix (i.e., the difference between the measurement X and the model Y as a function of factors G and F).

The elemental analysis of the measured samples as described in (Saraga et al., 2017) yielded 17 trace elements (Al, As, Ba, Cd, Co, Cr, Cs, Cu, Fe, Ga, Mn, Ni, Pb, Rb, Sr, V, Zn), 8 water-soluble ions (Cl^- , NO_3^- , SO_4^{2-} , Na^+ , NH_4^+ , K^+ , Mg^{2+} , Ca^{2+}), elemental and organic carbon (EC, OC). For avoiding double mass counting (K and K^+ , Ca and Ca^{2+}), the ionic forms were excluded from the analysis. In this study, a combined $\text{PM}_{2.5}$ and PM_{10} dataset was used as input to the PMF model. Eleven samples were initially excluded from the model: nine samples were excluded for too many missing species and two samples (one of each fraction) were treated as outliers and excluded from the analysis due to extremely high concentrations attributed to a single dust event recorded on May 4th, 2015. Co, As, Cd, Pb, Ga, Rb, Sr, Cs and Ba were set as ‘bad’ species and excluded from the analysis due to the high percentage of missing values. Ni, Zn, Cu, and Ca were set as ‘weak’ species due to their low signal/noise (S/N) ratio and/or bad scaled residuals (d-matrix). PM concentration was set as total variable. Concentration data

below the DL were substituted with half of the DL and missing concentration data were substituted with the median value for the specific specie (Polissar et al., 2001). Equation-based uncertainties were used (estimated based on analytical errors), and the modeling extra uncertainty was adjusted to 12%.

Constraints were applied in order to reduce rotational ambiguity and obtain more physically meaningful factors (Paatero et al., 2002). Bootstrap runs (BS), in addition to the advanced tools of the PMF (v.5), the displacement (DISP) and the bootstrap-displacement (BS-DISP) methods, were used to evaluate rotational ambiguity.

2.4 Estimation of Traffic Intensities

The principal road network of Doha city is comprised of 230 road segments, across the whole Doha Metropolitan Area, spanning an area of around 25 km diameter, according to the Doha's Public Work Authority (DPWA personal communication). Traffic intensities are measured regularly throughout the years using automatic and manual means, but not consistently in terms of frequency and location. For example, the DPWA carried out three major studies to measure traffic intensities at multiple roads in 2010, 2011, and 2015 for most of the road segments in Doha but not always the same. Their campaigns took place once or twice a year for a week (7 days) and collected hourly traffic counts per segment, lane, and type of vehicle (heavy duty and light duty). At the same time, the road network is changing drastically with new layouts and transitions from roundabouts to intersections and interchanges, while the total number of vehicles increased by 34% from 2011 to 2015. In addition, most of the main roads consist of multiple lanes per direction and with vehicle numbers approaching 100k per day. All these factors considered, reference traffic counts dataset was synthesized using the available data with

traffic counts per hour, day of the week, and type of vehicle (heavy and light duty). Although important, more details on the derivation of this dataset are beyond the scope of this study.

2.5 fPM Emissions from Barren Lands and Construction Sites

Emissions modelling of the fPM from barren lands and construction sites was based on two different approaches. The former is calculated based on the well-established and validated model of Schaap et al. (2009) for the regional modelling of PM. This model was developed on the principles of the proposed atmospheric dust cycle by Marticorena and Bergametti (1995). Later, others (e.g. Liora et al., 2015) elaborated on the parameters of these models and proposed improvements. For this study, we selected the original regional modelling scheme as a baseline with no later modifications. Calculation of fPM emissions from construction sites is based on a simpler approach but its parameters were developed and validated in our own field studies that characterized emissions from the local surface soil (i.e. Calcisols) (Hassan et al., 2016). More details can be found in the related literature.

For both approaches, detailed land use and land cover (LULC) information are required. For the current study area, i.e. State of Qatar, there were two relevant LULC databases. The first is the Harmonized World Soil Database (HWSD) (FAO, 2014) which provides surface soil (0-100 cm depth) data. The second is based on a recent local geological study (West and Al-Mulla, 2013). For this study, we combined information from these two databases with recent satellite photos (Google Earth) and site-visits to develop one LULC map and database that incorporates, as well, the exposed (barren) lands within the metropolitan area. The final map had a resolution of $0.5 \times 0.5 \text{ km}^2$ and includes the clay-sand-silt compositions for each relevant LULC category.

2.6 Dispersion Modelling of PM

The latest CALPUFF model (v7.2.1), coupled with the interface from Lakes Environmental Software was chosen to calculate the atmospheric dispersion of PM. CALPUFF is a non-steady state Lagrangian Gaussian puff model that has been developed since 1990 and employed in many similar studies ([Chatzimichailidis et al., 2014](#); [Holnicki et al., 2016](#)). The CALPUFF modelling system includes CALMET model for the post-processing and downscaling of mesoscale meteorological data, and CALPOST for post-processing of the results. The same configuration was used for both the traffic and non-traffic PM emissions. As a note, CALPUFF source code was modified to expand the hard-coded constraints on the number of total sources and released puffs that can be handled simultaneously. Also, CALPOST source code was modified to correct a bug concerning the time-series export of single-source contribution data. Either modified versions were compiled using the Lahey/Fujitsu Fortran v7.3 (Windows) and PGI Fortran v16.10 (Unix) and were verified using the originally compiled model with accuracy up to the inherent numerical error. Verification results are not shown here for brevity. Meteorological input was provided in the form of MM5 (Fifth-Generation Penn State/NCAR Mesoscale Model) weather data based on a parameterization described at an earlier work ([Gopalaswami et al., 2015](#)). The modelling domain was 200×200 km² centered on the city of Doha with a nested resolution following the LULC map resolution. An additional discrete receptor was added at the field campaign site. Finally, all simulations were executed using the high-performance computing (RAAD2) facilities at Texas A&M University at Qatar.

Eventually, dispersion modelling resulted in four discrete contributions to the PM concentration levels: (i) the contribution of barren lands and constructions sites, later referred to as (AR10), estimated using the fPM emissions described in Section 2.5, (ii) the contribution of

light-duty vehicles and (iii) the contribution of heavy-duty vehicles estimated using a unity emission factor (1 mg VKT^{-1} ; referred to later as (c_{ldv}) and (c_{hdv}), respectively), and (iv) the contribution of the road surface – Aeolian resuspension using again a unity emission factor (1 mg km^{-1} ; referred to later as c_{road}). A fifth dispersion contribution was estimated indirectly combining results from the PMF analysis and the AR10 contribution (VfPM) and further discussed in Section 3.2.

3. Results and Discussion

3.1 Measurements

During the campaign period, hourly air temperatures ranged from 17.4 to 45.1 °C, relative humidity from 5.9% to 34.0%, and wind speeds from 0.0 to 8.6 m s^{-1} , as demonstrated in the Supplementary Information (SI); Table S1. Calmer winds were observed between midnight and early mornings (mostly $<3 \text{ m/s}$) while higher averages up to 4.1 m s^{-1} were reported between 12:00 and 18:00h (local time). Obviously, the measured wind direction is biased by the nearby building, as this is reflected in the wind rose diagram (SI, Figure S1). Therefore, mesoscale modelling results have been employed after validation with airport wind data ($\sim 6 \text{ km SE}$). Thus, prevailing winds blew from North and East.

Table S2 in the SI presents the ambient mass concentrations as reported by the particle counter and estimated with the weighted filters. Due to the high variability of the PM measurements we categorized the PMCs measurements into typical days ($<200 \mu\text{g m}^{-3}$), minor dust events ($200\text{--}1000 \mu\text{g m}^{-3}$), and major dust events ($>1000 \mu\text{g m}^{-3}$) following recommendations from past studies (Al Dabbous, 2015; Draxler, 2001; Saraga et al., 2017). Noteworthy, the majority of the herein reported major dust events coincides with the respective

warnings issued by the Qatar Civil Aviation Authority, and were excluded from further analysis following the QA/QC criteria presented in Section 2.3.

Figure 3 presents the hourly time series of the measured PMCs and wind speeds. Missing data are due to technical issues or maintenance of the instruments. The pollution roses (Figure 4) present the measured PMCs partitioned by wind speed and wind direction (major events excluded). It appears that PM_{10} levels were mainly affected by sources to the East, which is consistent with the major road next to the building and also the coastline. In contrast, $PM_{2.5}$ levels appear to be affected by a “point” source to the Northeast that coincides with a large nearby road intersection. We believe that this difference between the $PM_{2.5}$ and PM_{10} pollution roses is attributed to the different driving conditions at the road intersection compared to the free flow, also reported in similar studies (Goel and Kumar (2015)), and the sea salt and shipping emissions coming from the coastline (see Section 3.2 on the source apportionment results).

A more detailed analysis of the diurnal and weekday patterns is shown in Figure 5, using a threshold of $PM_{10} < 200 \mu g m^{-3}$ (the suggested daily mean for non-dusty days). On average, PM_{10} followed the traffic patterns, it peaked in the late morning on weekdays (1000h-1200h) and again in the afternoon (at 1400h-1500h & 1800h), corresponding to the usual rush hours due to schools and offices activity. PM_{10} levels were slightly lower during weekends (Friday-Saturday), compared to weekdays (Sunday-Thursday) with the exception of Monday. It is probable that the patterns observed or not observed (e.g. the lowest levels should have been observed on Friday morning due to the close to zero traffic) were affected by low intense dust events. To explore this further, the threshold value for PM_{10} was lowered progressively to around $PM_{10} < 160 \mu g m^{-3}$, reaching to a PM_{10} weekday pattern closer to the one expected and without affecting the diurnal one. On the contrary, the $PM_{2.5}$ diurnal profile is “flat” with very small variations and the

weekday profile counterintuitive with lowest values observed on Tuesdays and the highest ones on Thursdays-Saturdays. The corresponding weekday pollution roses (with $\text{PM}_{10} < 160$ or $200 \mu\text{g m}^{-3}$) also identified the road intersection, in the North-Northeast, as a major hot spot with a few more hot spots in other directions, where other road intersection lies (see Figure 2). On the other hand, the pollution rose of Tuesdays' was the only one with the road intersection in the North-Northeast not being identified as a major hot spot (see Figure S2 in SI). Based on the rather short available data it was not possible to extract a definite cause for these observations, even though the temporal variation of the meteorological parameters (e.g. wind speed, temperature and humidity) was investigated for the whole 12 weeks of the field campaign. In any case, the $\text{PM}_{2.5}$ weekday variations were rather small ($\pm 10 \mu\text{g m}^{-3}$) and the $\text{PM}_{2.5}$ diurnal profile so "flat" that both the assumptions of a) significant sources with non-temporal variation (e.g. industry, construction) and b) counterbalancing sources and formation/suppression mechanisms (e.g. boundary layer height, humidity, secondary particles, normal traffic patterns, intense heavy-duty traffic during night) are equally plausible. Increasing the threshold value up to $\text{PM}_{10} < 1000 \mu\text{g m}^{-3}$ did not change the $\text{PM}_{2.5}$ patterns nor the PM_{10} diurnal variation, but flattened the PM_{10} weekday variation.

Figure 3 Time series of the hourly measured $PM_{2.5}$ and PM_{10} concentrations using the particle counter.

Figure 4 Pollution roses of the PMCs ($\mu g m^{-3}$) hourly measurements ($PM_{10} < 1000 \mu g m^{-3}$).

(a)

(b)

Figure 5 Mean diurnal and weekly variations of (a) $PM_{2.5}$ and (b) PM_{10} concentrations using two subsets of the data ($PM_{10} < 200 \mu g m^{-3}$ & $PM_{10} < 160 \mu g m^{-3}$). The shaded areas indicate the 95% confidence intervals.

3.2 Source Apportionment

For each factor 20 runs were performed, in order to obtain Q-value stability. All runs converged and Q values ranged between ± 1.2 %. Q-robust value was lower than 1.5 times the Q-true value, indicating that outliers are not significantly impacting the Q value. The optimal number of factors was determined by examining the Q values for PMF solutions resulting from a range of the -number of factors- values without excluding the solution's physical validity (Reff et al., 2007). A range of solutions were examined with different number of factors (3-12), but six was the maximum number of factors corresponding to meaningful sources.

Factors 1 and 3 were both associated with Na^+ and Mg^{2+} , which are typical tracers of sea salt. This is an expected result as the measuring site is located at a distance of ~ 1.2 km from the eastern coastline of Doha. Factor 1, however, was characterized as “aged sea salt” (ASS) due to the absence of chlorine and the presence of high level of NO_3^- , indicating the formation of sodium nitrate (Pérez et al., 2016). Factor 3 was characterized as “fresh sea salt” (FSS) due to the high loadings of Cl^- , Na^+ and to a lesser extent Mg^{+2} (Amato et al., 2016a), and the Cl^-/Na ratio of 1.63 (Pey et al., 2013). Due to the abundance of NH_4^+ and SO_4^{2-} , Factor 2 was characterized as “secondary aerosols” (SEA) which are typical components of regional and long-range transport emissions (Saraga et al., 2019). Factor 4 was the dominant factor with 29.2% contribution and showed mixed composition of crustal elements (Ca, Al, Fe and Mn) as well as brake wear markers (Zn and Cu). This mixture depicts a profile of dust source that possibly includes non-exhaust from traffic (VfPM), due to the abrasion of vehicle wear and dust resuspension (RESUS) (Heo et al., 2017). Factor 5 was strongly associated with carbonaceous elements EC and OC, and to a lesser extent Cu and Zn, with an OC/EC ratio of 0.86, falling within the ranges previously reported for (VEX) emissions (Amato et al., 2016a). Factor 6 was

identified as a combined combustion factor of “fuel oil/shipping” (FO/SHP) emissions. The fuel oil combustion is owed to the presence of V, Ni, and SO_4^{2-} possibly from industrial processes (Sowlat et al., 2013), which would be a reasonable guess giving that an industrial area is located ~ 13.1 km away from the monitoring site. V and Ni are also tracers of shipping emissions. Although the estimated V/Ni ratio (1.58) in this study was less than the ranges previously reported for shipping emissions (Pandolfi et al., 2011), the V/Cu ratio (8.43) is $\gg 1$ which, according to Tolis et al. (2015), is a value of clear primary shipping emissions. The percentage contributions and profiles of the factors are shown in Figure 6.

In the present study, two constraints needed to be applied with the limitation of dQ% being kept at the lowest value of 0.5%: i) for the RESUS factor: Ca and Fe were pulled up maximally and ii) for the VEX factor: EC was pulled up maximally. Compared to the initial results, no substantial changes to either the constrained profiles or the constrained contributions were observed, amplifying the accuracy of the selected solution. A third constraint was applied to the FO/SHP as it showed no OC content, which should not be the case for fuel combustion. OC was pulled up maximally for the FO/SHP factor and minimally for ASS factor. The constrained solution showed similar convergence as the base run and yielded a dQ% of 0.41, and improved the solution with OC content of 6.13% for the subject factor. After the constraints application, BS, DIS and BS-DIS runs were conducted for evaluating the model's solutions. In particular, 100 bootstrap runs were performed (default value of minimum correlation 0.6), and the results were regarded reliable as >90% of the factors were mapped. DIS runs resulted in the validity of the final PMF solutions as no factor swaps (i.e. exchanges) were observed for the smallest dQ_{max}=4 while the decrement of Q was <1%. Through BS-DISP analysis, the final solution was considered to be valid as <0.5% drop was associated with the Q value (Norris et al., 2014).

416

(a)

(b)

Figure 6 Factor contributions (constrained) to the total PM and the elemental profiles for each of the PMF factors as percentage contributions and absolute concentrations (ASS: aged sea salt,

SEA: secondary aerosol, FSS: fresh sea salt, RESUS: resuspension, VEX: vehicle exhaust, FO/SHP: fuel oil/shipping)

3.3 Derived Emission Factors

Following the presented methodology, three distinct sets of data have been created: (i) the six PMF factors (identified PM sources), (ii) four meteorological variables (including the estimated friction velocity), and (iii) five specific contributions including the light-duty, heavy-duty, road surface and PM area sources (barren land and construction sites) that have been derived employing dispersion modelling. The symbols and the correlation matrix for the 15 aforementioned daily datasets are presented in Figure 7. Although many interesting arguments could be extracted from this matrix, the focus of this study is the vehicle-induced PM (VEX and VfPM). PMF identified the exhaust (VEX) PM which is positively correlated with the vehicle (0.51-0.52) and road (0.53) dispersion contributions. On the other hand, the resuspension factor (RESUS) is poorly correlated with the traffic-related contributions, and moderately correlated with the dispersion contributions (AR10) from barren lands and construction sites revealing that this factor includes airborne crustal material beyond the road dust. Removing this contribution from the resuspension factor (RESUS-AR10), the non-exhaust (VfPM) dispersion contribution is derived. The VfPM has no correlation with the traffic related contributions indicating no linear relation with the contributions. It is worth noting here that VEX is negatively correlated with the wind speed (ws) which implies that strong winds dilute the impact of exhaust traffic PM rather than increasing them. In fact, the very strong winds have not been included in the source apportionment analysis following the exclusion of the major dust events. Finally, both VEX and VfPM show a negative correlation with humidity, indicating the effect of moisture in suppressing traffic emissions.

Figure 7 Correlation heatmap for the estimated PMF factors (ASS: aged sea salt, SEA: secondary aerosol, FSS: fresh sea salt, RESUS: resuspension, VEX: traffic, FO/SHP: fuel oil/shipping, and Total), meteorological quantities (ws: wind speed, T: temperature, ustar: friction velocity, and humidity), and dispersion modelled quantities (AR10: barren land and construction, VfPM: RESUS-AR10, c_ldv: contribution of light-duty vehicles, c_hdv: contribution of heavy-duty vehicles, and road: contribution of road surface).

The derivation of the EFs [mg VKT^{-1}] for the VEX and VfPM was achieved using linear and non-linear regression analysis (with single and multi-variables) offered by the Stats package (v3.5.2 “lm” and “nls”) in R software. In both cases (VEX and VfPM), it was not possible to

derive separate PM EFs for the heavy and light duty vehicle types. This was due to the low counts of heavy-duty vehicles for the roads close to the measurement's location, although all 230 road segments have been employed. Therefore, we decided to apply four different combinations of the traffic counts in the regression analysis and use the results to estimate the range of the proposed EFs: (i) total contributions of traffic ($c_{ldv}+c_{hdv}$), (ii) only the light-duty contributions (c_{ldv}), (iii) an arbitrary 50% increase in heavy-duty emission factor ($c_{ldv}+1.5 c_{hdv}$), and (iv) an arbitrary 100% increase in heavy-duty emission factor ($c_{ldv}+2 c_{hdv}$), compared to light duty.

As a result, the estimated $EF(PM_{10})$ for VEX ranged from 620 to 730 $mg\ VKT^{-1}$ (Adjusted $R^2 \sim 0.84$; Figure 8a).

Figure 8 Scatter plot examples of the measured vs predicted (a) exhaust (VEX) and (b) non-exhaust (VfPM) PM_{10} concentration levels with the proposed emission factors. Vectors correspond to the daily wind speed and prevailing direction, and colors the average relative humidity [%]. The outer dense-dashed lines represent the factor of two (FAC2) range (1:2 and 2:1), the blue dashed line is the regression line with the blue shade corresponding to the 95% confidence intervals.

Concerning the estimated EF(PM₁₀) for VfPM, they ranged from 1080 to 1410 mg VKT⁻¹ (Adjusted R² ~ 0.70; Figure 8b). All EFs previously reported secured a confidence level (p-value for the t-test) of 99% or better.

Further to the above and following past literature suggestions, we performed a non-linear regression analysis to derive the VfPM EF using road and meteorological parameters. Among the many combinations tested, the below EF models for non-exhaust PM₁₀ showed good agreement (i.e. adjusted R² ~ 0.70-0.71), but not better than the linear model:

$$EF = a \times u^b$$

$$EF = a \times u^b + cH$$

$$EF = (a + cH) \times u^b$$

where, EF [mg VKT⁻¹] is the emission factor, u is the wind speed [m s⁻¹], H is the humidity [%], and the a , b and c are empirical parameters. At the same time, the significance of each new parameter was rather weak (p-value in the range of 60%-80%). Conversely, based on the data and analysis of this study, a linear model for the VfPM EF is proposed.

4. Conclusions

In this study we derived one of the first EFs and emission models to estimate VEX and VfPM in areas with arid desert climates. A combination of field measurements, chemical characterization, and dispersion modelling was used to calculate emission contributions at the measuring site. Following a linear regression analysis (using solely the traffic contributions), the fitted PM₁₀ EF for VEX and VfPM ranged from 620 to 730 mg VKT⁻¹ (adjusted R² ~ 0.84), and 1080 to 1410 mg VKT⁻¹ (adjusted R² ~ 0.70), respectively. A non-linear regression analysis provided an emission model for VfPM having similar performance to the linear model, but increased the p-value indicating a weak hypothesis. The EFs presented herein seem to be higher

than the ranges reported in Europe (Amato et al., 2016b; Ketzel et al., 2007) and within the ranges reported in USA (Abu-Allaban et al., 2003). Although the results are plausible, we recommend further investigations to improve emissions modelling of VfPM and build a diverse database for the arid desert climates. For example, future field campaigns should cover a wider and longer range of seasons, include multiple streets with different traffic patterns, consider the road surface dust-load, incorporate gaseous-pollutants as tracers of exhaust emissions, and further validate the available meteorological data.

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