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Review article

A systematic review of land use regression models for volatile organic compounds

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GRAPHICAL ABSTRACT

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ABSTRACT

Various aspects of land use regression (LUR) models for volatile organic compounds (VOCs) were systematically reviewed. Sixteen studies were identified published between 2002 and 2017. Of these, six were conducted in Canada, five in the USA, two in Spain, and one each in Germany, Italy, and Iran. They were developed for 14 different individual VOCs or groupings: benzene; toluene; ethylbenzene; *m*-xylene; *p*-xylene; (*m*/*p*)-xylene; *o*-xylene; total BTEX; 1,3-butadiene; formaldehyde; *n*-hexane; total hydro carbons; styrene; and acrolein. The models were based on measurements ranging from 22 sites in El Paso (USA) to 179 sites in Tehran (Iran). Only four studies in Rome (Italy), Sabadell (Spain), Tehran, and Windsor (Canada) met the Cocheo's criterion of having at least one passive sampler per 3.4 km^2 of study area. The range of R^2 values across all models was from 0.26 for 1,3-butadiene in Dallas (USA) to 0.93 for benzene in El Paso. The average R^2 values among two or more studies of the same VOCs were as follows: benzene (0.70); toluene (0.60); ethylbenzene (0.66); (*m*/*p*)-xylene (0.65); *o*-xylene (0.61); total BTEX (0.66); 1,3-butadiene (0.46); and formaldehyde (0.56). The common spatial predictors of studied VOC concentrations were dominated by traffic-related variables, but they also included proximity to ports in the USA, number of chimneys in Canada, altitude in Spain, northern latitudes in Italy, and proximity to sewage treatment plants and to gas filling stores in Iran. For the traffic-related variables, the review suggests that large buffers, up to 5,000 m, should be considered in large cities. Although most studies reported logical directions of association for predictors, some reported inconsistent results. Some studies included log-transformed predictors while others divided one variable by another. Only six studies provided the p-values of predictors. Future work may incorporate chemistry-transport models, satellite observations, meteorological variables, particularly temperature, consider specific sources of aromatic vs aliphatic compounds, or may develop hybrid models. Currently, only one national model has been developed for Canada, and there are no global

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LUR models for VOCs. Overall, studies from outside North America and Europe are critically needed to describe the wide range of exposures experienced by different populations.

1. Introduction

Approximately 8% of global deaths and 4% of global disability-adjusted life-years (DALYs) were attributed to ambient air pollution in 2015, making it one of the leading modifiable risk factors for the Global Burden of Disease (GBD) worldwide (Cohen et al., 2017). These estimates were based on exposure to particulate matter ≤ 2.5 μm ($PM_{2.5}$) and ozone (O_3) (Brauer et al., 2016), for ischemic heart disease (IHD), cerebrovascular disease (ischemic stroke and hemorrhagic stroke), lung cancer, chronic obstructive pulmonary disease (COPD), and lower respiratory infections (LRI) (Cohen et al., 2017). The GBD estimates would be considerably higher if more air pollutants and health outcomes where included in the analyses (Amini et al., 2014a). For example, pollutants such as nitrogen dioxide (NO_2) and air toxics have been independently associated with increased risk of morbidity and mortality from the health outcomes already considered (Cesaroni et al., 2013; Filippini et al., 2015; Samoli et al., 2006; Thomas et al., 2014). On the other hand, health outcomes such as leukemia and other cancers (Lavigne et al., 2017; Weichenthal et al., 2017), neurodegenerative diseases (Chen et al., 2017; Heydarpour et al., 2014), and many others have been associated with the air pollutants (Bakian et al., 2015; Künzli et al., 2000; Nhung et al., 2017; Thurston et al., 2017; West et al., 2016).

The GBD estimates are partly influenced by the estimated relative risks extracted from cohort studies on long-term exposure to air pollution (GBD 2015 Risk Factors Collaborators, 2016). The most commonly used exposure assessment method in health outcomes analysis has been land use regression (LUR) (Hoek et al., 2013). Since its introduction in Europe (Briggs et al., 1997), the approach has been extensively used over the last 20 years to estimate the spatial variability in a wide range of pollutants, mainly in high-income countries (HICs) (Beelen et al., 2013, 2014; Dirgawati et al., 2016; Eeftens et al., 2012; Henderson et al., 2007; Raaschou-Nielsen et al., 2013). However, there have also been some studies in low- and middle-income countries (LMICs) (Amini et al., 2014b, 2016; Gurung et al., 2017; Lee et al., 2017; Yang et al., 2017).

Generally, LUR models have been applied to map concentrations of particulate matter and nitrogen oxides and, to a lesser extent, other pollutants, such as volatile organic compounds (VOCs) (Hoek et al., 2008). There are hundreds of VOCs species in ambient air, but they are all characterized by a low boiling point and ready transformation to the gaseous phase (Monks et al., 2009). Important groups of VOCs include: aliphatic alkanes, such as hexane; aromatic alkylbenzenes, such as benzene, toluene, ethylbenzene, and xylenes (BTEX); halogenated hydrocarbons, such as tetrachloromethane; and terpenes, such as carene (Baldasano et al., 1998). To date, most LUR models of VOCs have focused on the aromatic alkylbenzenes, which are ubiquitous in fossil fuels and combustion products (Monks et al., 2009).

One of the largest studies to use LUR for long-term exposure assessment in research on the development of chronic disease is the European Study of Cohorts for Air Pollution Effects (ESCAPE) (Beelen et al., 2013; Eeftens et al., 2012). However, studies such as ESCAPE and the GBD have not used LUR to model VOCs, despite their carcinogenicity (International Agency for Research on Cancer (IARC), 2016; Lipfert, 2017). We see three possible reasons for this. First, most studies in HICs have reported very low concentrations of VOCs in the ambient air (Guerreiro et al., 2014). Second, both $PM_{2.5}$ and O_3 have been consistently useful for predicting the range of health outcomes included in large studies (Brauer et al., 2016; Cohen et al., 2017). Third, study of VOCs has not been a priority for funding agencies, likely due to the first and second points. However, some studies have reported that ambient VOC concentrations are very high within large LMICs cities where large numbers of people are exposed (Amini et al., 2017a; Hoque et al., 2008; Matysik et al., 2010).

There is no evidence of a safe threshold for carcinogenic VOCs (e.g., benzene), and they may contribute to a considerable burden of disease even at concentrations below the current standards (Beelen et al., 2014; Künzli et al., 2015; Kutlar Joss et al., 2017). As such, the exclusion of VOCs from burden of disease studies likely leads to underestimation of the deaths and DALYs attributable to a wide range of cancers, particularly in densely populated and highly exposed LMICs. Based on the evidence to date, one key difference between VOCs and pollutants such as $PM_{2.5}$ or O_3 is the distributions of their ambient concentrations in HICs and LMICs. The burden of disease associated with $PM_{2.5}$ and O_3 also tends to be high in LMICs, but the vast literature from HICs can be used to inform LMICs calculations because there is considerable overlap between the distributions across contexts (Brauer et al., 2016). When it comes to VOCs, however, there appears to be little overlap in the distributions, which is consistent with the nature of pollutants that disperse quickly to the atmosphere. This results in a dearth of literature from highly-studied areas that can be used to inform calculations for highly-exposed areas.

Given (1) the potential burden of disease attributable to VOCs and (2) the limited information about population exposures to VOCs, we have conducted a systematic review of the published LUR models. Many different components of LUR modeling, such as monitoring data, geographic predictors, model development, and validation have been reviewed elsewhere (Hoek et al., 2008; Jerrett et al., 2005; Ryan and LeMasters, 2007), but no other study has focused on VOCs. In this article, we aim to provide a systematic review of LUR models developed for VOCs over the last two decades to summarize different elements of this literature, including: the geographic distribution of published studies; the VOCs modeled; the number of measurement sites used for VOC modeling; air quality data collection methods; VOC pollutant-specific predictor variables; and model evaluation. Finally, we discuss the knowledge gaps and future directions of research in this area.

2. Materials and methods

We searched 12 databases in *The Web of Science*: *Web of Science™ Core Collection*; *Medline®*; *BIOSIS Citation Index℠*; *BIOSIS Previews®*; *Current Contents Connected®*; *Data Citation Index℠*; *Derwent Innovations Index℠*; *Inspect®*; *KCI-Korean Journal Database; Russian Science Citation Index; SciELO Citation Index; and Zoological Record®*. The search phrases and keywords were: Land use regression; LUR; volatile organic compound; VOC; BTEX; benzene; toluene; and xylene. There was no restriction on timespan or language, and the final search was done on August 25, 2017. Only original research articles were retained for the analyses. Studies were only included if they were related to air pollution and they had developed an LUR model for at least one VOC. Studies were excluded if they had only applied VOC estimates from an LUR model described in another publication, or if no full-text was available (e.g. a conference abstract). The references of identified LUR articles on VOCs were also checked to identify any relevant articles missed by the search.

The full-text of all identified articles was reviewed and the following data were extracted: the year of publication; citation; study location (country, city/area, and population size); modeled VOCs; years of measurements; measurement method; the number of distributed sites used to build the LUR; the number of reference sites; median measured

Table 1
Summary of 16 LUR studies published for VOCs up to August 25, 2017.

Country (citation)	City/Domain	Population (Million)	Modeled area (km^2)	# of measured sites	Cocheo's minimum required # of sites[a]	Measurement period	Data source: # of measurement campaigns	# of reference sites (measurement period)[b]	Modeled VOCs[c]	Median (min–max) measured benzene ($\mu g/m^3$)
Canada (Wheeler et al., 2008)	Windsor, Ontario	0.2	146	54	43	Feb, May, Aug, and Oct 2004	Passive sampling: four 2-week campaigns	N/A	B, T	0.9 (0.5–1.4)
Canada (Atari and Luginaah, 2009)	Sarnia	0.07	165	39	48	Oct 2005	Passive sampling: one 2-week campaign	N/A	B, T, E, *mp*-X, *o*-X, TBTEX	0.93 (0.3–3.4)
Canada (Su et al., 2010)	Toronto	2.7	630	50	183	Jul 25–Aug 9, 2006	Passive sampling: one 2-week campaign	N/A	B, *n*-H, TH	0.6 (0.4–1.3)
Canada (Hystad et al., 2011)	National	35	9 million	53	N/A	Annual averages for 2006	Fixed-site monitoring data	N/A	B, E, 1,3-B	N/A
Canada (Oiamo et al., 2015)	Ottawa	0.9	2790	42	809	Oct 7–21, 2008 and May 6–20, 2009	Passive sampling: two 2-week campaigns	N/A	B, T, *mp*-X	0.5 (0.3–0.8)
Canada (Poirier et al., 2015)	Halifax	0.3	70% of Halifax population	50	N/A	Oct 20–Nov 3, 2010 and Jan 5–Jan 19, 2011	Passive sampling: two 2-week campaigns	N/A	B, T	0.4 (0.3–2.6)
USA (Smith et al., 2006)	El Paso	0.6	664	22	193	Nov & Dec 1999	Passive sampling: two 1-week campaigns	N/A	B	2.5 (1.6–4.9)
USA (Mukerjee et al., 2009)	Detroit	0.7	370	25	107	Summer 2005	Passive sampling: one 5-week campaign	N/A	B, T, E, *mp*-X, *o*-X, TBTEX, S, 1,3-B	1.5 (1.1–2.2)
USA (Johnson et al., 2010)	New Haven	0.1	52	XX[e]	N/A	Jul–Aug 2001	Pseudo-observations: estimates of modeling based on CMAQ and AERMOD	N/A	B	1.1 (N/A)
USA (Smith et al., 2011)	Dallas	1.3	999	24	290	Aug 1–Sep 5, 2006	Passive sampling: one 5-week campaign	N/A	B, T, E, *o*-X, *mp*-X, 1,3-B	0.7 (0.3–1.3)
USA (Kheirbek et al., 2012)	New York	8.4	789	69[f]	229	March 22–Jun 01, 2011	Passive sampling: five 2-week campaigns	3 (~2-months)	B, TBTEX, F	0.8 (0.3–2.3)
Spain (Aguilera et al., 2008)	Sabadell	0.2	38	55	11	Apr 2005–March 2006	Passive sampling: four 1-week campaigns	N/A	TBTEX	0.9 (0.4–3.1)
Spain (Fernandez-Somoano et al., 2011)	Asturias	0.1	483	67	140	Jun 2005 and Nov 2005	Passive sampling: two 1-week campaigns	N/A	B	2 (0.04–9.2)
Germany (Carr et al., 2002)	Munich	1.4	310	34	90	Dec 1996–Feb 1998	Passive sampling: twelve 4-week campaigns	N/A	B, T, E	N/A[g] (1.8–14.5)
Italy (Gaeta et al., 2016)	Rome[d]	N/A	64	43	19	May 31–Jun 14, 2011 and Jan 11–25, 2012	Passive sampling: two 2-week campaigns	1 (N/A)	B, T, A, F	2.2 (1.6–2.6)
Iran (Amini et al., 2017b)	Tehran	9	613	179	178	Apr 2015–May 2016	Passive sampling: three 2-week campaigns	5 (~1-year)	B, T, E, *p*-X, *m*-X, *o*-X, TBTEX	7.8 (2.1–25.8)

[a] Cocheo's minimum required number of sites = 0.29 × study area (in km^2) (Cocheo et al., 2008).
[b] Reference sites are used for temporal variation adjustment in passive sampling campaigns.
[c] Abbreviations or letters denote: B (benzene), T (toluene), E (ethylbenzene), *p*-X (*p*-xylene), *m*-X (*m*-xylene), *o*-X (*o*-xylene), *mp*-X (*m/p*-xylene), TBTEX (total benzene, toluene, ethylbenzene, and xylenes), *n*-H (*n*-hexane), TH (total hydrocarbons), 1,3-b (1,3-butadiene), S (styrene), F (formaldehyde), and A (acrolein).
[d] This LUR study has been conducted in the Ciampino Airport area, Rome, Italy.
[e] This study used up to 285 sites for LUR modeling. However, there has been no real measurement and they were pseudo-observations from a hybrid modeling based on CMAQ and AERMOD (Isakov et al., 2012; Johnson et al., 2010).
[f] In this study 14 sites were measured at each campaign resulting in about 70 sites over five consecutive 2-week campaigns.
[g] Site-specific statistics are reported. Median (min-max) for benzene were at 18 traffic sites 8.7 (4.5–14.5) and at 16 school sites 2.6 (1.8–3.8).

Table 2
Details of 16 LUR studies published for VOCs up to August 25, 2017.

Authors/citation City (Country) # of sites	Modeled VOC	Median (min – max) (μg/m^3)	Model R^2/Adj. R^2	Validation method	Validation R^2	Predictors (direction of association (positive; negative; or N/A)); (p-value)
Carr et al. (Carr et al., 2002) Munich (Germany) 34	Benzene	18 traffic sites: 8.7 (4.5–14.5) 16 school sites: 2.6 (1.8–3.8)	0.80/N/A	N/A	N/A	- Traffic counts within 0–50 m (N/A); (N/A) - Traffic counts within 0–300 m (N/A); (N/A) - Traffic counts with high traffic jam percentages (N/A); (N/A)
	Toluene	18 traffic sites: 28.5 (16.3–44.3) 16 school sites: 9.9 (6.0–14.5)	0.76/N/A	N/A	N/A	same as benzene (N/A); (N/A)
	Ethylbenzene	18 traffic sites: 5.6 (3.1–9.0) 16 school sites: 1.9 (1.2–2.7)	0.79/N/A	N/A	N/A	same as benzene (N/A); (N/A)
Smith et al. (Smith et al., 2006) And Mukerjee et al. (2012) (Mukerjee et al., 2012) El Paso (USA) 22 schools	Benzene	2.5 (1.6–4.9)	0.93/N/A	N/A	N/A	- Population density within census block group or set radii (positive); (N/A) - Point source (categorical or continuous) (negative if categorical/positive if continuous); (N/A) - Distance to nearest international border crossing (negative); (N/A)
Aguilera et al. (Aguilera et al., 2008) Sabadell (Spain) 55	Ln(Total BTEX)	17.5 (3.5–34.1)	0.74/N/A	- LOOCV[a] - Leave 15% out	No LOOCV validation R2 reported	- Altitude (negative); (N/A) - Distance to nearest major road (negative); (N/A) - Distance to nearest secondary road (negative); (N/A) - Distance to nearest parking lot (negative); (N/A)
Wheeler et al. (Wheeler et al., 2008) Windsor (Canada) 54	Benzene	0.9 (0.5–1.4)	0.73/N/A	-Leave five sites out from modeling	0.78	- Length of major roads within 100 m (positive); ($p < 0.001$) - Length of expressways and primary highways within 50 m (positive); ($p < 0.001$) - Detroit VOC emission point sources within 4,000 m (positive); ($p < 0.001$) - Windsor VOC emission point sources within 3,000 m (positive); ($p < 0.001$)
	Toluene	2.7 (1.3–6.3)	0.46/N/A	same	0.65	- Distance to Ambassador Bridge (negative); ($p < 0.001$) - Length of major roads within 200 m (positive); ($p = 0.055$) - Length of primary highways within 100 m (positive); ($p = 0.033$) - Windsor VOC emission point sources within 1,000 m (positive); ($p < 0.001$)
Atari et al. (Atari and Luginaah, 2009) Sarnia (Canada) 39	Benzene	0.93 (0.3–3.4)	0.78/0.76	-LOOCV -Leave 10%, 20%, and 50% out	0.75–0.81	- Industry 1,600 m (positive); (N/A) - Dwelling 1,200 m (positive); (N/A) - Highway 800 m (positive); (N/A)
	Toluene	2.6 (0.8–6.9)	0.81/0.79	same	0.77–0.86	- Industry 2,800 m (positive); (N/A) - Open 600 m (negative); (N/A) - Highway 800 m (positive); (N/A)
	Ethylbenzene	0.5 (0.1–1.1)	0.81/0.79	same	0.79–0.86	- Industry 2,600 m (positive); (N/A) - Dwelling 1,400 m (positive); (N/A) - Highway 800 m (positive); (N/A)
	(*m/p*) xylene	1.2 (0.4–2.8)	0.80/0.78	same	0.78–0.81	- Industry 1,600 m (positive); (N/A) - Dwelling 1,200 m (positive); (N/A) - Highway 800 m (positive); (N/A)
	o -xylene	0.5 (0.1–1.2)	0.80/0.78	same	0.77–0.79	- Industry 1,600 m (positive); (N/A) - Dwelling 1,200 m (positive); (N/A) - Highway 800 m (positive); (N/A)

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Table 2 (*continued*)

Authors/citation City (Country) # of sites	Modeled VOC	Median (min – max) (μg/m³)	Model R^2/Adj. R^2	Validation method	Validation R^2	Predictors (direction of association (positive; negative; or N/A)); (p-value)
	Total BTEX	5.7 (1.8–14.5)	0.81/0.79	same	0.80–0.84	- Industry 2,500 m (positive); (N/A) - Dwelling 1,400 m (positive); (N/A) - Highway 900 m (positive); (N/A)
Mukerjee et al. (2009) (Mukerjee et al., 2009) And Mukerjee at al. (2012) (Mukerjee et al., 2012) Detroit (USA) 25 schools	Ln(Benzene)	1.5 (1.1–2.2)	0.43/N/A	Leave two sites out from modeling	N/A	- Distance to nearest medium traffic road (negative); (N/A) - Traffic intensity within 1,000 m (negative); (N/A) - Population density within 500 m (positive); (N/A) - Distance(m) to nearest large Manganese emission source (negative); (N/A) - Distance to nearest international border crossing (positive); (N/A)
	Ln (Toluene)	6.10 (0.4–8.6)	0.31/N/A	same	N/A	- Distance(m) to nearest large Manganese emission source (negative); (N/A) - Distance to nearest international border crossing (positive); (N/A)
	Ln (Ethylbenzene)	0.8 (0.5–1.6)	0.63/N/A	same	N/A	- Log-distance to nearest medium traffic road (negative); (N/A) - Distance to nearest high traffic road (negative); (N/A) - Log-traffic intensity within 1,000 m of location (negative); (N/A) - Distance (m) to nearest large VOC emission source (negative); (N/A) - Log(distance to nearest large Manganese emission source) (negative); (N/A) - Distance to nearest border crossing (positive); (N/A)
	Ln (*m*/*p*-xylene)	2.7 (1.6–5.3)	0.55/N/A	same	N/A	- Distance to nearest medium traffic road (negative); (N/A) - Distance to nearest high traffic road (negative); (N/A) - Traffic intensity within 1,000 m of location (negative); (N/A) - Log(distance) to nearest large VOC emission source (negative); (N/A) - Distance to nearest large PM2.5 emission source (positive); (N/A) - Log(distance to nearest large Manganese emission source) (negative); (N/A) - Distance to nearest international border crossing (negative); (N/A)
	Ln (*o*-xylene =	0.90 (0.5–1.5)	0.60/N/A	same	N/A	- Log-distance to nearest medium traffic road (negative); (N/A) - Distance to nearest high traffic road (negative); (N/A) - Log-population density within 500 m (positive); (N/A) - Distance to nearest large VOC emission source (negative); (N/A) - Log-distance to nearest large PM2.5 emission source (positive); (N/A) - Log(distance to nearest large Manganese emission source) (negative); (N/A) - Distance to nearest international border crossing (positive); (N/A)
	Ln (Total BTEX)	2.81 (1.95–4.11) in ppb units	0.40/N/A	same	N/A	- Distance to nearest medium traffic road (negative); (N/A)

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Table 2 (*continued*)

Authors/citation City (Country) # of sites	Modeled VOC	Median (min – max) (μg/m^3)	Model R^2/Adj. R^2	Validation method	Validation R^2	Predictors (direction of association (positive; negative; or N/A)); (p-value)
						- Log(distance to nearest large Manganese emission source) (negative); (N/A)
	Ln (styrene)	0.1 (0.1–0.3)	0.43/N/A	same	N/A	- Distance to nearest moderate traffic road (positive); (N/A) - Distance to nearest high traffic road (negative); (N/A) - Traffic intensity within 1,000 m of location (positive); (N/A) - Population density within 500 m (positive); (N/A) - Distance(m) to nearest large Manganese emission source (negative); (N/A) - Distance to nearest international border crossing (negative); (N/A)
	Ln (1,3-butadiene)	3.5 (2.5–6.5)	0.43/N/A	same	N/A	- Log-distance to nearest moderate traffic road (negative); (N/A) - Traffic intensity within 1,000 m of location (positive); (N/A) - Population density within 500 m (positive); (N/A) - Log-distance to nearest large VOC emission source (positive); (N/A) - Log-distance to nearest large PM2.5 emission source (negative); (N/A) - Distance(m) to nearest large Manganese emission source (negative); (N/A) - Distance to nearest international border crossing (positive); (N/A)
Johnson et al. (Johnson et al., 2010) New Haven (USA) 25–285[b]	Benzene	1.1 (N/A – N/A)	N/A/ 0.67–0.89	-LOOCV -Hold out 33 to 293 sites	No LOOCV validation R2 reported; Hold-out R2 = 0.21–0.71	- Traffic intensity (N/A); (N/A) - Proximity to roadways (N/A); (N/A) - Proximity to ports of harbors (N/A); (N/A) - Proximity to industrial sources (N/A); (N/A)
Su et al. (Su et al., 2010) Toronto (Canada) 50	Ln(Benzene)	0.6 (0.4–1.3)	0.67/N/A	bootstrap (in total 18 bootstrap models applied)	No bimodal shape was presented for predictors coefficients	- Expressway in 100 m (positive); (N/A) - Major road in 50 m (positive); (N/A) - Commercial land use in 2,900 m (positive); (N/A) - Industrial in 1,200 m (positive); (N/A)
	Ln(*n*-hexane)	1.0 (0.7–3.3)	0.68/N/A	same	same	- Local road in 50 m (negative); (N/A) - Industrial in 1,200 m (positive); (N/A) - Open land use area in 50 m (negative); (N/A) - Number of chimneys in 2050 m (positive); (N/A) - Population Density in 1,150 m (positive); (N/A) - Soil brightness in 1,650 m (positive); (N/A)
	Ln(Total hydrocarbons)	25.6 (17.1–77.5)	0.66/N/A	same	same	- Industrial in 1,200 m (positive); (N/A) - Open land use area in 50 m (negative); (N/A) - Open land use area in 1,200 m (negative); (N/A) - Number of chimneys in 2050 m (positive); (N/A) - Soil brightness in 1,650 m (positive); (N/A)
Fernandez-Somoano et al. (Fernandez-Somoano et al., 2011) Asturias (Spain) 67	Ln(Benzene)	2 (0.04–9.2)	0.73/N/A	LOOCV	0.71	- Altitude (negative); (N/A) - Continuous urban land cover in 300 m (negative); (N/A) - Agriculture/forest land cover in 300 m (negative); (N/A) - Distance to nearest road (positive); (N/A) - Discontinuous urban land cover in 1,000 (positive); (N/A)
Hystad et al. (Hystad et al., 2011) National (Canada) 53	Benzene	N/A (N/A – N/A)	0.62/N/A	LOOCV Bootstrap	0.12; Normal distribution for predictors coefficients	- Major road length in 10 km (positive); ($p < 0.001$) - National Pollutant Release Inventory (NPRI) emissions in 10 km (positive); ($p < 0.001$)
	Ethylbenzene	N/A (N/A – N/A)	0.67/N/A	same		- Population in 10 km (positive); ($p < 0.001$)

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Table 2 (*continued*)

Authors/citation City (Country) # of sites	Modeled VOC	Median (min – max) (μg/m^3)	Model R^2/Adj. R^2	Validation method	Validation R^2	Predictors (direction of association (positive; negative; or N/A)); (p-value)
					Normal distribution for predictors coefficients	- NPRI emissions in 2 km (positive); ($p < 0.001$)
	1,3-Butadiene	N/A (N/A – N/A)	0.68/N/A	same	Normal distribution for predictors coefficients	- Road length in 750 m (positive); ($p < 0.001$) - Highway in 500 m (positive); ($p = 0.002$) - Commercial land use area in 10 km (positive); ($p = 0.01$)
Smith et al. (Smith et al., 2011) Dallas (USA) 24 fire stations (models are for summer and winter, respectively)	Benzene	0.7 (0.3–1.3)	0.72/N/A	N/A	N/A	- Distance to nearest road with 10,000 < traffic volume < 20,000 vehicles per day (negative); (N/A) - Distance to nearest road with 70,000 < traffic volume < 80,000 vehicles per day (negative); (N/A) - Distance to nearest road with 100,000 < traffic volume < 120,000 vehicles per day (positive); (N/A) - Traffic intensity within 1,000 m (positive); (N/A)
	Benzene	1.2 (0.8–1.7)	0.49/N/A	N/A	N/A	- Distance to nearest road with 10,000 < traffic volume < 20,000 vehicles per day (positive); (N/A) - Distance to nearest road with 70,000 < traffic volume < 80,000 vehicles per day (negative); (N/A) - Traffic intensity within 1,000 m (positive); (N/A)
	Ln (Toluene)	2 (0.6–4.3)	0.41/N/A	N/A	N/A	- Distance to nearest road with 70,000 < traffic volume < 80,000 vehicles per day (negative); (N/A) - Distance to nearest road with 100,000 < traffic volume < 120,000 vehicles per day (negative); (N/A) - Distance to source with NOx emissions > 570,000 lbs per year (negative); (N/A) - Distance to source with 21,000 < NOx emissions < 221,000 lbs per year (positive); (N/A)
	Ln (Toluene)	2.3 (0.9–6.6)	0.41/N/A	N/A	N/A	- Distance to nearest road with 70,000 < traffic volume < 80,000 vehicles per day (negative); (N/A) - Distance to nearest road with 100,000 < traffic volume < 120,000 vehicles per day (negative); (N/A) - Distance to source with NOx emissions > 570,000 lbs per year (negative); (N/A) - Distance to source with 21,000 < NOx emissions < 221,000 lbs per year (negative); (N/A)
	Ln (Ethylbenzene)	0.4 (0.1–0.8)	0.63/N/A	N/A	N/A	- Distance to nearest road with 70,000 < traffic volume < 80,000 vehicles per day (negative); (N/A) - Distance to nearest road with 100,000 < traffic volume < 120,000 vehicles per day (negative); (N/A) - Traffic intensity within 1,000 m (negative); (N/A) - Distance to source with benzene emissions > 270,000 lbs per year (negative); (N/A) - Distance to source with NOx emissions > 570,000 lbs per year (positive); (N/A) - Distance to source with 21,000 < NOx emissions < 221,000 lbs per year (negative); (N/A)
	Ln (Ethylbenzene)	0.4 (0.2–1.3)	0.40/N/A	N/A	N/A	- Distance to nearest road with 70,000 < traffic volume < 80,000 vehicles per day (negative); (N/A) - Distance to nearest road with 100,000 < traffic volume < 120,000 vehicles per day (negative); (N/A) - Traffic intensity within 1,000 m (positive); (N/A) - Distance to source with benzene emissions > 270,000 lbs per year (negative); (N/A) - Distance to source with NOx emissions > 570,000 lbs per year (positive); (N/A)

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Table 2 (*continued*)

Authors/citation City (Country) # of sites	Modeled VOC	Median (min – max) (μg/m^3)	Model R^2/Adj. R^2	Validation method	Validation R^2	Predictors (direction of association (positive; negative; or N/A)); (p-value)
	Ln (*o*-xylene)	0.4 (0.1–0.9)	0.46/N/A	N/A	N/A	- Distance to source with 21,000 < NOx emissions < 221,000 lbs per year (negative); (N/A) - Distance to nearest road with 70,000 < traffic volume < 80,000 vehicles per day (negative); (N/A) - Distance to nearest road with 100,000 < traffic volume < 120,000 vehicles per day (negative); (N/A) - Distance to source with benzene emissions > 270,000 lbs per year (negative); (N/A) - Distance to source with NOx emissions > 570 000 lbs per year (positive); (N/A)
	Ln (*o*-xylene)	0.4 (0.2–1.2)	0.37/N/A	N/A	N/A	- Distance to nearest road with 70,000 < traffic volume < 80,000 vehicles per day (negative); (N/A) - Distance to nearest road with 100,000 < traffic volume < 120,000 vehicles per day (negative); (N/A) - Distance to source with NOx emissions > 570,000 lbs per year (negative); (N/A)
	Ln(*m*/*p*-xylene)	1.1 (0.4–2.7)	0.71/N/A	N/A	N/A	- Distance to nearest road with 10,000 < traffic volume < 20,000 vehicles per day (positive); (N/A) - Distance to nearest road with 70,000 < traffic volume < 80,000 vehicles per day (negative); (N/A) - Distance to nearest road with 100,000 < traffic volume < 120,000 vehicles per day (positive); (N/A) - Traffic intensity within 1,000 m (positive); (N/A) - Distance to source with benzene emissions > 270 000 lbs per year (negative); (N/A) - Ln (Distance to source with ethylbenzene emissions > 4,400 lbs per year) (positive); (N/A) -Distance to source with NOx emissions > 570,000 lbs per year (positive); (N/A)
	Ln(*m*/*p*-xylene)	1.1 (0.4–3.9)	0.40/N/A	N/A	N/A	- Distance to nearest road with 10,000 < traffic volume < 20,000 vehicles per day (positive); (N/A) - Distance to nearest road with 70,000 < traffic volume < 80,000 vehicles per day (negative); (N/A) - Distance to nearest road with 100,000 < traffic volume < 120,000 vehicles per day (positive); (N/A) - Traffic intensity within 1,000 m (positive); (N/A) - Distance to source with benzene emissions > 270,000 lbs per year (negative); (N/A) - Ln (Distance to source with ethylbenzene emissions > 4,400 lbs per year) (positive); (N/A) - Distance to source with NOx emissions > 570,000 lbs per year (positive); (N/A)
	Ln(1,3-butadiene)	3.6 (1.9–7.5)	0.26/N/A	N/A	N/A	- Distance to nearest road with 70,000 < traffic volume < 80,000 vehicles per day (negative); (N/A) - Distance to nearest road with 100,000 < traffic volume < 120,000 vehicles per day (negative); (N/A) - Distance to source with benzene emissions > 270,000 lbs per year (negative) - Distance to source with 21,000 < NOx emissions < 221,000 lbs per year (negative)
	Ln(1,3-butadiene)	5.8 (2.4–15.7)	0.40/N/A	N/A	N/A	- Distance to nearest road with 70,000 < traffic volume < 80,000 vehicles per day (negative); (N/A)

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Table 2 (*continued*)

Authors/citation City (Country) # of sites	Modeled VOC	Median (min – max) (μg/m^3)	Model R^2/Adj. R^2	Validation method	Validation R^2	Predictors (direction of association (positive; negative; or N/A)); (p-value)
Kheirbek et al. (Kheirbek et al., 2012) New York City (USA) 67–69 (14 sites at each campaign)	Benzene	0.8 (0.3–2.3)	0.65/N/A	Build model by 85% of observations and test the remaining 15% for validation	0.62	- Distance to nearest road with 100,000 < traffic volume < 120,000 vehicles per day (negative); (N/A) - Distance to source with benzene emissions > 270 000 lbs per year (negative); (N/A) - Distance to source with 21,000 < NOx emissions < 221,000 lbs per year (negative); (N/A) - Number of traffic signals within 400 m (positive); ($p < 0.001$) - Length of interstate, state, and county highways within 100 m (positive); ($p < 0.001$) - Reference site mean (positive); ($p = 0.022$)
	Total BTEX	4.7 (1.5–20.4)	0.70/N/A	same	0.65	- Number of signals within 450 m (positive); ($p < 0.001$) - Kernel-weighted smooth of solvent-based industry locations in 500 m (positive); ($p < 0.001$) - Reference site mean (positive); ($p = 0.01$)
	Formaldehyde	2.2 (1.2–3.7)	0.83/N/A	same	0.68	- Reference site mean (positive); ($p < 0.001$) - Number of signals within 400 m (positive); ($p < 0.001$) - Road length within 100 m (positive); ($p < 0.001$) - Built space within 100 m (positive); ($p = 0.001$)
Oiamo et al. (Oiamo et al., 2015) Ottawa (Canada) 41–42	Benzene	0.5 (0.3–0.8)	0.78/N/A	Bootstrap	Stable predictors coefficients	- Population in 2,500 m (positive); ($p < 0.001$) - Length of highway in 300 m (positive); ($p < 0.01$) - NPRI VOC facility count in 4 km (positive); ($p < 0.01$) - Distance to National Pollutant Release Inventory VOC facility (negative); ($p < 0.05$) - Intersection count in 100 m (positive); ($p < 0.01$)
	Toluene	1.8 (0.5–5.1)	0.79/N/A	same	One variable was unstable	- Area of parks/open space in 1,200 m (negative); ($p < 0.01$) - Length of highway in 350 m (positive); ($p < 0.01$) - National Pollutant Release Inventory VOC facility count in 4 km (positive); ($p < 0.01$) - National Pollutant Release Inventory toluene facility count in 8 km (positive); ($p < 0.05$) - Intersection count in 100 m (positive); ($p < 0.05$)
	m/p -xylene	0.7 (0.2–1.3)	0.75/N/A	same	One variable was unstable	- Area of parks/open space in 1,400 m (negative); ($p < 0.001$) - Length of highway in 350 m (positive); ($p < 0.01$) - Intersection count in 100 m (positive); ($p < 0.01$) - National Pollutant Release Inventory VOC facility count in 4 km (positive); (N/A)
Poirier et al. (2015) (Poirier et al., 2015) Halifax Regional Municipality (Canada) 50	Benzene	0.4 (0.3–2.6)	0.61/N/A	N/A	N/A	N/A
	Toluene	0.4 (0.2–1.1)	0.63/N/A	N/A	N/A	N/A
Gaeta et al. (Gaeta et al., 2016) The Ciampino Airport, Rome (Italy) 39–43	Benzene	2.2 (1.6–2.6)	0.57/0.53	LOOCV	0.50	- The North latitude (positive); ($p < 0.05$) - Product of traffic intensity of the nearest road and the inverse of distance to the nearest road (positive); ($p < 0.05$) - Number of inhabitants in a 100 m buffer (positive); ($p < 0.05$)
	Toluene	5.8 (3.4–7.3)	0.54/0.50	same	0.40	- The North latitude (positive); ($p < 0.05$)

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Authors/citation City (Country) # of sites	Modeled VOC	Median (min – max) ($\mu g/m^3$)	Model R^2/Adj. R^2	Validation method	Validation R^2	Predictors (direction of association (positive; negative; or N/A)); (p-value)
						- Product of traffic intensity of the nearest road and the inverse of distance to the nearest road (positive); ($p < 0.05$) - Number of inhabitants in a 100 m buffer (positive); ($p < 0.05$)
	Acrolein	2.8 (2.6–4.1)	0.55/0.51	same	0.50	- Number of inhabitants in a 100 m buffer (positive); ($p < 0.05$) - Traffic intensity of the nearest major road (positive); ($p < 0.05$) - Traffic intensity of the nearest road (positive); ($p < 0.05$) - Aircraft contribution to HC concentration (positive); ($p < 0.05$)
	Formaldehyde	2.7 (2.4–2.9)	0.29/0.24	same	0.13	- The North latitude (negative); ($p < 0.05$) - Number of inhabitants in a 500 m buffer (positive); ($p < 0.05$) - Traffic intensity of the nearest major road (positive); ($p < 0.05$)
Amini et al. (Amini et al., 2017b) Tehran (Iran) 179	Ln (Benzene)	7.8 (2.1–25.8)	0.70/0.68	LOOCV	0.66	- All road (5,000) (positive); ($p < 0.02$) - Distance to sewage treatment plants (negative); ($p < 0.02$) - Ancillary roads (50) (positive); ($p < 0.02$) - Highways (50) (positive); ($p < 0.02$) - Taxi lines (25) (positive); ($p < 0.02$) - Log-distance to taxi lines (negative); ($p < 0.02$) - Distance to all bus terminals (negative); ($p < 0.02$) - Sensitive land use areas (700) (negative); ($p < 0.02$) - Official/commercial land use areas (2,500) (positive); ($p < 0.02$)
	Ln (Toluene)	23.2 (6.1–88.9)	0.65/0.64	same	0.60	- All road (50) (positive); ($p < 0.02$) - Log-distance to alleys (positive); ($p < 0.02$) - Log-distance to taxi lines (positive); ($p < 0.02$) - Log-distance to gas filling stores (positive); ($p < 0.02$) - Official/commercial land use areas (2000) (positive); ($p < 0.02$) - All road (5,000) (positive); ($p < 0.02$) - Product of green space areas in buffer 5,000 m/distance to green spaces (negative) - Distance to sewage treatment plants (negative); ($p < 0.02$) - Sensitive land use areas (2000) (negative); ($p < 0.02$)
	Ln (Ethylbenzene)	5.7 (1.4–9.8)	0.66/0.64	same	0.61	- Taxi lines (25) (positive); ($p < 0.05$) - Log-distance to taxi lines (negative); ($p < 0.05$) - Official/commercial land use areas (2000) (positive); ($p < 0.05$) - Residential land use areas (3,500) (positive); ($p < 0.05$) - All road (5,000) (positive); ($p < 0.05$) - Product of green space areas in buffer 5,000 m/distance to green spaces (negative); ($p < 0.05$) - Distance to sewage treatment plants (negative); ($p < 0.05$) - Industrial land use areas (5,000) (positive); ($p < 0.05$) - Sensitive land use areas (500); ($p < 0.05$) - Highways (50) (positive); ($p < 0.05$)

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Table 2 (*continued*)

Authors/citation City (Country) # of sites	Modeled VOC	Median (min – max) (μg/m^3)	Model R^2/Adj. R^2	Validation method	Validation R^2	Predictors (direction of association (positive; negative; or N/A)); (p-value)
						- All road (50); ($p < 0.05$)
	Ln (*p*-xylene)	5.6 (1.7–16.8)	0.64/0.62	same	0.59	- Taxi lines (50) (positive); ($p < 0.02$) - Ancillary roads (50) (positive); ($p < 0.02$) - Product of highways in buffer 50 m/distance to highways (positive); ($p < 0.02$) - Green space land use areas (5,000) (negative); ($p < 0.02$) - Official/commercial land use areas (2000) (positive); ($p < 0.02$) - Distance to all bus parking areas (negative); ($p < 0.02$) - All road (5,000) (positive); ($p < 0.02$) - Distance to sewage treatment plants (negative); ($p < 0.02$) - Product of green space areas in buffer 5,000 m/distance to green spaces (negative); ($p < 0.02$) - Distance to urban facilities land use areas (positive); ($p < 0.02$) - Arid or undeveloped land use areas (700) (negative); ($p < 0.02$) - Sensitive land use areas (400) (negative); ($p < 0.02$)
	Ln (*m*-xylene)	10.4 (2.6–34.7)	0.66/0.64	same	0.61	- Ancillary roads (50) (positive); ($p < 0.05$) - Highways (50) (positive); ($p < 0.05$) - Taxi lines (25) (positive); ($p < 0.05$) - Log-distance to taxi lines (negative); ($p < 0.05$) - Official/commercial land use areas (2000) (positive); ($p < 0.05$) - Distance to sewage treatment plants (negative); ($p < 0.05$) - All road (5,000) (positive); ($p < 0.05$) - Industrial land use areas (3,500) (positive); ($p < 0.05$) - Distance to all bus terminals (negative); ($p < 0.05$) - Urban facilities land use (3,500) (negative); ($p < 0.05$) - Sensitive land use areas (2000) (negative); ($p < 0.05$)
	Ln (*o*-xylene)	6.1 (2.1–17.8)	0.66/0.63	same	0.59	- Taxi lines (50) (positive); ($p < 0.05$) - Ancillary roads (50) (positive); ($p < 0.05$) - Blocks (250) (positive); ($p < 0.05$) -Transportation land use areas (5,000) (positive); ($p < 0.05$) - Official/commercial land use areas (3,500) (positive); ($p < 0.05$) - Green space land use areas (5,000) (negative); ($p < 0.05$) - Sensitive land use areas (200) (negative); ($p = 0.067$) - Product of green space areas in buffer 5,000 m/distance to green spaces (negative); ($p < 0.05$) - Distance to sewage treatment plants (negative); ($p < 0.05$) - Product of taxi lines in buffer 3,500 m/distance to taxi lines (positive); ($p = 0.066$) - Distance to agricultural land use areas (positive); ($p < 0.05$) - Product of alleys in buffer 5,000 m/distance to alleys (positive); ($p < 0.05$)

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concentrations (min – max); important model diagnostics including R^2, adjusted R^2, validation method, cross-validation R^2; and the predictive spatial variables included in the final LUR model, along with their buffer sizes (where applicable), direction of the effect, and p-values (if available).

3. Results and discussion

The initial search identified 39 records. One conference abstract that overlapped with one original research article was immediately removed. After further assessments of the remaining records, 23 more were excluded according to the study criteria. Checking the references of the remaining publications resulted in identification of an early VOCs model in Munich (Germany) where the authors did not use the term LUR despite using LUR methods (Carr et al., 2002).

Overall, we identified 16 LUR studies for VOCs published between 2002 and 2017. Of these, 11 were conducted in North America including six in Canada (Atari and Luginaah, 2009; Hystad et al., 2011; Oiamo et al., 2015; Poirier et al., 2015; Su et al., 2010; Wheeler et al., 2008) and five in the USA (Johnson et al., 2010; Kheirbek et al., 2012; Mukerjee et al., 2009, 2012; Smith et al., 2006; Smith et al., 2011). Of the remainder, two were conducted in Spain (Aguilera et al., 2008; Fernandez-Somoano et al., 2011), one in Germany (Carr et al., 2002), one in Italy (Gaeta et al., 2016), and one in Iran (Amini et al., 2017b). The populations of the cities covered by the VOC models ranged from ~70,000 in Sarnia (Canada) (Atari and Luginaah, 2009) to ~9 million in Tehran (Iran) (Amini et al., 2017b). A national model for Canada estimated VOCs for the whole population of the country, which is ~35 million people. The modeled areas also ranged widely, from 38 km^2 in Sabadell (Spain) (Aguilera et al., 2008) to 2,790 km^2 in Ottawa (Canada) (Oiamo et al., 2015) (Table 1).

The 16 studies developed LUR models for VOCs based on measurements ranging from 22 sites in El Paso (USA) (Smith et al., 2006) to 179 sites in Tehran (Iran) (Amini et al., 2017b). There was also one study in New Haven (USA) where benzene concentrations were simulated at 285 sites using a hybrid of two air quality modeling systems (CMAQ and AIRMOD), and the LUR models were built using the pseudo-observations (Johnson et al., 2010) (Table 2). Although there is no consensus in the LUR community about the number of sites needed to develop the spatial models (Amini et al., 2014b; Hoek et al., 2008), studies have shown that using a small number of sites may lead to overestimates of model performance. One study in Girona (Spain) found that > 80 sites were needed across 46 km^2 to adequately capture the range of NO_2 concentrations and to properly estimate long-term spatial variability (Basagaña et al., 2012). Authors further recommended that > 100 sites might be needed for LUR models of NO_2 in more complex urban settings (Basagaña et al., 2012). Beckerman et al. (2008) reported that NO_2 concentrations near an expressway were strongly correlated with benzene (0.85) and total hydrocarbons (0.74), but less correlated with toluene (0.63), ethylbenzene (0.51), *m*/*p*-xylene (0.46), and *o*-xylene(0.51) (Beckerman et al., 2008). Thus, we assume that a similar number of sites might be needed for VOCs as for NO_2. In a related study, Cocheo et al. (2008) evaluated how the number of passive BTEX samplers could be reduced in four European cities (Dublin, Madrid, Paris and Rome) while maintaining the quality of the results achieved with a larger number of sites. They found that each 3.4 km^2 in urban areas needed at least one passive sampler, and recommended that the number of sites needed can be calculated by $0.29 \times A$ where A denotes the study area in km^2 (Cocheo et al., 2008). Overall, 10 out of 16 studies in our review failed to meet this criterion, including those in Sarnia, Toronto, national model of Canada, Ottawa, El Paso, Detroit, Dallas, New York, Asturias, and Munich. However, the studies in Windsor, Sabadell, Rome, and Tehran well met this criterion (Table 1). The national Canadian study developed models based on 53 sites (Hystad et al., 2011) for a 9.9 million km^2 modeling domain. The study in Dallas (USA) used 22 sites in an area of 664 km^2 (Smith et al., 2006),

Table 2 (*continued*)

Authors/citation City (Country) # of sites	Modeled VOC	Median (min – max) ($\mu g/m^3$)	Model R^2/Adj. R^2	Validation method	Validation R^2	Predictors (direction of association (positive; negative; or N/A)); (p-value)
	Ln (Total BTEX)	58.6 (16.4–195.0)	0.66/0.64	same	0.61	- Distance to all bus parking areas (negative); ($p < 0.05$) - Highways (250) (positive); ($p < 0.05$) - Distance to educational areas (positive); ($p < 0.05$) - Taxi lines (50) (positive); ($p < 0.05$) - Product of highways in buffer 50 m/distance to highways (positive); ($p < 0.05$) - Log-distance to official/commercial land use areas (negative); ($p < 0.05$) - Log-distance to gas filling stores (negative); ($p < 0.05$) - Official/commercial land use areas (2000) (positive); ($p < 0.05$) - Urban facilities land use (200) (negative); ($p = 0.06$) - Distance to sewage treatment plants (negative); ($p < 0.05$) - Product of green space areas in buffer 5,000 m/distance to green spaces (negative); ($p < 0.05$) - All road (5,000) (positive); ($p < 0.05$) - Sensitive land use areas (2000) (negative); ($p < 0.05$) - Ancillary roads (50) (positive); ($p < 0.05$)

[a] LOOCV, leave one out cross-validation.
[b] Note that these sites were pseudo-observations not real measurements.

and the study in New York City (USA) used 69 sites for an area of 789 km^2 (Kheirbek et al., 2012). Only the study in Tehran had > 100 measurement sites available, with 179 sites for an area of 613 km^2 (Amini et al., 2017b).

Three of the studies modeled only benzene, and four modeled all BTEX species. Others modeled a few different VOCs, such as *n*-hexane, 1,3-butadiene, styrene, formaldehyde, and acrolein (Table 1). Given that the BTEX species have been highly correlated (Amini et al., 2017a; Hoque et al., 2008), future studies might consider restricting measurements and models to benzene, given its well-established adverse health effects. However, studies have shown that modeling each VOC separately might help to identify pollutant-specific predictor variables and/or sources, which could inform air pollution control programs (Amini et al., 2017b). In addition, some VOCs such as acetaldehyde and acetone have shown low to moderate correlations with other VOCs because their sources can be different (Possanzini et al., 2007; von Schneidemesser et al., 2010), so separate models will be needed in some cases.

Passive sampling was used to collect data for 14 of the 16 studies (Table 1). The Canadian national study used routinely collected fixed-site monitoring data, and the aforementioned study in New Haven used pseudo-observations based on air quality simulations (Table 1). Most studies have used inexpensive passive samplers because regulatory networks (1) may not measure VOCs routinely and (2) are unlikely to capture the whole range and spatial variability of concentrations across the study area. These considerations are especially true for LMICs, where missing data, could be challenging to impute, making long-term averages nearly impossible to estimate for LUR (Amini et al., 2014b). Previous studies have demonstrated that passive samplers provide accurate data compared with other types of measurements (Marc et al., 2015; Stevenson et al., 2001). Furthermore, the hourly benzene pseudo-observations simulated by Johnson et al. (2010) were compared with measurements at one site, and the agreement was within a factor of two (Johnson et al., 2010). As such, the uncertainty of LUR models based on simulated data might be larger than the uncertainly of models based on true measurements.

The measurement periods for the passive sampling campaigns have been: (1) one 2-week campaign in two studies; (2) one 5-week campaign in two studies; (3) two 1-week campaigns in two studies; (4) four 1-week campaigns in one study; (5) two 2-week campaigns in three studies; (6) three 2-week campaigns in one study; (7) four 2-week campaigns in one study; (8) five 2-week campaigns in one study; and (9) twelve 4-week campaigns in one study. Finally, the national Canadian model used annual averages at 53 fixed regulatory monitoring over one entire year, and the study in New Haven simulated the benzene data over a single summer (Table 1). While data from regulatory networks can provide high temporal coverage, they cannot adequately capture spatial variability on air pollutant concentrations (Kanaroglou et al., 2005). Simulated data can capture both temporal and spatial trends with high resolution, but their accuracy is uncertain in the absence of a sufficient number of measurements for robust evaluation.

Although the ideal approach in passive sampling campaigns is to measure at all sites throughout the study period (e.g. annually), this might not be feasible due to budget and/or logistic constraints (Amini et al., 2017a). Therefore, the investigators usually measure at a small number of sites throughout the year, which we refer to as reference sites. The long-term means for the distributed sites are then adjusted based on their comparison with the reference site(s) using various approaches (Amini et al., 2017a; Eeftens et al., 2012). Again, there is no consensus in the LUR community about the number of reference sites that should be measured, the number of measurement periods, the length of those periods, the number of campaigns needed, or how measurements should be spaced in time. From review of the available literature, these choices often depend on the study domain, local meteorology, and geographic characteristics.

Previous studies have shown that measurements from reference sites are needed to obtain robust estimates of the long-term mean at the distributed sites (Amini et al., 2017a). However, only three of the 16 studies used reference sites in their analyses (Table 1). The study in New York City had three reference sites and the measurements were taken over five consecutive 2-week periods. This study used the raw concentrations from the distributed sites as the response variable for the LUR, and the reference site mean was used predictive covariate, along with the spatial variables (Kheirbek et al., 2012). The study in Rome had one reference site, but it was in 7 km removed from the distributed sites and it was not clear whether the site ran throughout the study or how the data were used to adjust the other measurements (Gaeta et al., 2016). The study in Tehran measured five reference sites for 25 2-week periods and adjusted the models by using the ratios of the measurements at distributed sites to concurrent levels at reference sites (Amini et al., 2017a, 2017b).

Fifteen of the 16 studies were conducted in areas where long-term benzene concentrations were lower than 5 $\mu g/m^3$, which is the standard value set by the European Union (Marco and Bo, 2013). Only one study in the highly polluted megacity of Tehran reported annual average concentrations above this threshold (Amini et al., 2017b). Otherwise, the maximum long-term mean concentrations of benzene was reported in El Paso, Texas, where one site had an annual average of 2.5 $\mu g/m^3$ (Mukerjee et al., 2012). The median long-term measured benzene concentrations at individual sites ranged from 0.5 $\mu g/m^3$ in Ottawa, which has a population of approximately 900,000 people, to 7.8 $\mu g/m^3$ in Tehran, which has a population of approximately 9 million people (Table 1). The low concentrations in North America are consistent with the fact that emissions of VOCs have been drastically reduced in HICs over the last two decades. One study reported reductions of −3% to −26% per year from 1998 to 2008 for some individual VOCs in London (von Schneidemesser et al., 2010). There are no long-term standards for ambient exposure to VOCs other than benzene, which made further comparison between studies more challenging. All in all, the available evidence suggests that VOCs have high concentrations in LMICs communities (Amini et al., 2017a; Hoque et al., 2008; Matysik et al., 2010) compared with HICs.

Overall, 15 out of the 16 studies modeled at least benzene. These models explained a range of variability in measured concentrations from 43% in Detroit to 93% in El Paso. The high value for El Paso was likely a function of the high number of model parameters ($n = 16$) relative to the small number of sites used in the analyses ($n = 22$) (Hoek et al., 2008). The average (standard deviation) R^2 across all 15 studies on benzene was 0.70 (0.12). In general the most important variables in the LUR models for benzene were indicators of traffic, such as population density, highway density, distance to major roads, and some distinct variables relevant to local conditions. Specifically, the common predictors were: (1) population density in six studies; (2) length of expressways and highways within a buffer in five studies; (3) distance to nearest major road in five studies; (4) industry within a buffer or distance to industries in three studies; (5) VOC emissions sources in three studies; (6) commercial land use in two studies; (7) altitude in two studies; (8) and distance to nearest international border in two studies. Many individual studies also included variables that did not appear or were not calculated for other studies, including: traffic counts within a buffer; intersection count within a buffer; number of traffic signals within a buffer; total length of roads within in buffer; taxi lines within a buffer; dwellings within a buffer; proximity to ports of harbor; northern latitudes; distance to sewage treatment plants; and distance to nearest bus terminals (Table 2).

Model evaluation was also varied across the studies and, once again, there is no consensus in the LUR community about the best methods. Leave one out cross-validation (LOOCV) has been the popular approach when the number of measurement sites is small (Amini et al., 2014b, 2016). However, one NO_2 study in the Netherlands reported that models based on a smaller number of sites ($n = 24$ in this case) performed poorly in hold-out external validation (Wang et al., 2012). The LUR validation methods used in the reviewed studies were: LOOCV in Asturias, Rome, and Tehran; leave two out in Detroit; leave five out in Windsor; leave out proportions ranging from 10% to 50% in Sarnia, Sabadell, New Haven, and New York City. Bootstrapping was used (sometimes in addition to LOOCV) for the national Canadian model, and the Canadian cities of Toronto and Ottawa (Table 2). No clear validation method was reported for the models in Munich (Carr et al., 2002), El Paso (Smith et al., 2006), Dallas (Smith et al., 2011), or Halifax (Poirier et al., 2015). The validation R^2 values ranged from 12% in national Canadian model (Hystad et al., 2011) to 81% in Sarnia (Atari and Luginaah, 2009) (Table 2). Overall, there is a critical need for studies that compare different LUR validation methods for all pollutants, and specifically for VOCs.

We identified nine LUR models for toluene, explaining a range of variability from 31% in Detroit to 81% in Sarina. The average (SD) R^2 of these nine models was 0.60 (0.18), and the predictor variables were similar to those for benzene. In addition to the general traffic surrogates, specific variables that described toluene concentrations were: (1) distance to the Ambassador Bridge in Windsor; (2) distance to nearest large Manganese emissions source in Detroit; (3) industrial land use in Sarnia; (4) northern latitude in Rome; (5) log-distance to gas filling stores in Tehran; and (6) distance to the nearest sewage treatment plant in Tehran. The cross-validation R^2 were 40% in Rome, 60% in Tehran, 65% in Windsor, and 77%–86% in Sarnia. The other five studies did not report cross-validation values. The validation method in Ottawa was bootstrapping, and one variable was identified as unstable in the model (Table 2).

Six of the 16 studies modeled ethylbenzene, with an average (SD) variability explained of 0.66 (0.13), ranging from 0.40 to 0.63 in Dallas to 81% in Sarnia. Once again, the predictors were similar to those used for modeling benzene and toluene. The cross-validation R^2 was 0.61 in Tehran, and ranged from 0.79 to 0.86 in seasonal models of Sarnia. No cross-validation R^2 was reported for the models in Munich, Detroit, Dallas, or national model of Canada (Table 2).

Five of the 16 studies modeled xylenes, including *m*-xylene, *p*-xylene, (*m/p*)-xylene, and *o*-xylene. The LUR models for (*m/p*)-xylene explained 0.40 to 0.70 of variability in the seasonal data for Dallas and up to 0.80 of the variability in Sarnia. The Tehran study modeled *m*- and *p*-xylene separately and explained 0.66 of variability in both pollutants. The average (SD) R^2 was 0.65 (0.14) for all studies on (*m/p*)-xylene, and the predictive variables were similar to those used for benzene. Only the study in Sarnia reported a validation R^2 for (*m/p*)-xylene, which ranged from 0.78 to 0.81. The validation R^2 in Tehran was 0.61 for *m*-xylene and 0.59 for *p*-xylene. The LUR models for *o*-xylene explained 0.37 to 0.60 of variability in the seasonal data for Dallas, and up to 0.80 in Sarnia with an average (SD) of 0.61 (0.15) for all studies. The validation R^2 for *o*-xylene was 0.59 in Tehran, and ranged from 0.77 to 0.79 in Sarnia (Table 2).

While different studies covered benzene, toluene, ethylbenzene, and xylenes separately, five of the studies summarized in the paragraphs above also modeled total BTEX. The average (SD) R^2 value for these models was 0.66 (0.16), ranging from 0.40 in Detroit to 0.81 in Sarnia. Once again, the spatial predictors of total BTEX were similar to those for the benzene models. The reported validation R^2 values were 0.61 in Tehran, 0.65 in New York City, and 0.80 to 0.84 in Sarnia (Table 2).

Beyond the BTEX species, three studies modeled 1,3-butadiene, explaining 26% of variability in Dallas, 43% in Detroit, and 68% in the national Canadian model. No validation R^2 has been reported for these models but the national Canadian model reported a normal distribution for predictors coefficients as they used bootstrap method for validation (Table 2). Two modeled formaldehyde with R^2 values of 0.29 in Rome and 0.83 in New York City. The validation R^2 was 0.13 in Rome and 0.68 in New York City (Table 2). One study modeled styrene in Detroit and could explain 43% of variability. One study in Rome modeled acrolein near an Italian airport with an R^2 value of 55%. In addition to traffic-related predictors, a variable called "aircraft contribution to hydrocarbon concentration" (spray estimates) was a significant predictor for acrolein (Table 2). Finally, one study modeled *n*-hexane and total hydrocarbons in Toronto, explaining 68% and 66% of variability, respectively. In addition to traffic-related variables, these models also included the number of chimneys and soil brightness. However, local road had a negative effect on *n*-hexane. The models were validated by bootstrap and reported no bimodal shape for predictor coefficients (Table 2).

In general, we observed discrepancy among studies regarding the direction of association for predictors. Although most of the studies reported logical directions (e.g., increasing benzene concentrations with increasing traffic intensity), some reported inconsistent results (Fernandez-Somoano et al., 2011; Mukerjee et al., 2012; Smith et al., 2011). The studies in Munich and Halifax were missing important information on variable direction, and could not be included in this assessment. Regarding the non-linear terms in the models, some studies included log-transformed predictors (Mukerjee et al., 2009; Smith et al., 2011) while others divided one variable by another variable (Amini et al., 2017b). Only 6 out of 16 studies provided the p-values of predictors for their models. Of those predictors, majority had p-value < 0.05 but all had < 0.1, which is acceptable in the LUR community (Amini et al., 2014b).

4. Conclusions

In this article we used a systematic approach to provide an overview of all LUR models that have been developed for VOCs. Generally, the important VOC predictors were traffic-related variables. However, other significant predictors included proximity to ports of harbor in USA, number of chimneys in Canada, altitude in Spain, northern latitudes in Italy, and proximity to sewage treatment plants, taxi lines, bus terminals, or gas filling stores in Iran. Many of the traffic-related and other variables used large buffers, up to 5,000 m, which may be important for describing ambient VOCs in large cities (Amini et al., 2017b). On the other hand, future studies may need to critically evaluate specific local characteristics and sources of each VOC to achieve the best possible models. They may consider the inclusion of satellite derived variables, incorporating meteorological variables, particularly for temperature, or may develop hybrid models. So far, only one LUR study for VOCs has incorporated the meteorological variables in the modeling process, but they were not selected for the final models (Amini et al., 2017b).

So far only one set of national models has been developed for VOCs in Canada, and the performance of the models was relatively good (Hystad et al., 2011). A recent study showed that LUR models can be developed globally when the authors built an NO_2 using 5220 air monitors in 58 countries and reported an R^2 of 0.54 (Larkin et al., 2017). Currently, there is no such model for VOCs, mainly due to scarce global measurements. It is possible that the simulation approach used by Johnson et al. (2010) or recent advancements in satellite observation of VOCs (Zhu et al., 2017), could provide one pillar needed for construction of VOC models at the national and global scales. The availability of such models would facilitate epidemiologic studies on VOCs even in areas where no measurements have been made.

As shown, the majority of LUR models have been developed in high income countries for the aromatic alkylbenzene group of VOCs where they include toxic pollutants. Further studies on VOCs from outside North America and Europe are critically needed to describe the wide range of exposures experienced by different populations and possible health effects in LMICs.

Conflicts of interest

Vahid Hosseini declares that he is affiliated to Tehran Air Quality Control Company (AQCC). The views expressed in this manuscript are those of the authors and do not necessarily reflect the views or policies of the Tehran AQCC. The rest of authors declare that they have no actual or potential financial competing interests.

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