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6/4/2025

City of Burnet City Council
1001 Buchanan Drive, Suite 4
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Burnet, TX 78611

Re: Opposition to Conditional Use Permit for Proposed Truck Stop near 104 County Road 108

Dear Mayor and Council Members,

I am writing to express my strong opposition to the proposed conditional use permit for a truck stop on the north corner of County Road 108 in Burnet, Texas. As a concerned resident, I believe that the proposed development does not meet the criteria outlined in **Section 118-64(e)** of the City of Burnet's Code of Ordinances governing conditional use permits and that it poses a serious threat to public health and the surrounding environment.

According to **Section 118-64(e) - General Criteria applicable to all conditional use permits**, a proposed use must not:

1. **Adversely affect the health or safety of persons residing or working in the vicinity of the proposed use.**
2. **Be detrimental to the public welfare or injurious to property or improvements in the neighborhood.**
3. **Be inconsistent with the goals, objectives, and policies of the comprehensive plan or applicable area plans.**

The proposed truck stop fails to meet these standards, particularly in light of the nearby **assisted retirement living facility currently under construction at 104 County Road 108**, located within 300 feet of the proposed site. This facility will house one of the most vulnerable segments of our population—older adults—who are especially susceptible to airborne toxins such as **benzene**, a known carcinogen emitted from gas station operations.

Health Concerns:

Research indicates that **benzene and other harmful pollutants** are emitted from underground fuel storage tank vents and during vehicle refueling. While some states require protective vapor recovery systems, **Texas does not mandate such protections**, thereby increasing the exposure risk to nearby residents and vulnerable populations. Scientific studies have detected benzene emissions at distances of **up to 524 feet** from gas stations (PMC7020915), and the **U.S. EPA**

advises caution when siting schools within 1,000 feet of gas stations, a precaution that should extend to senior housing and other residential areas.

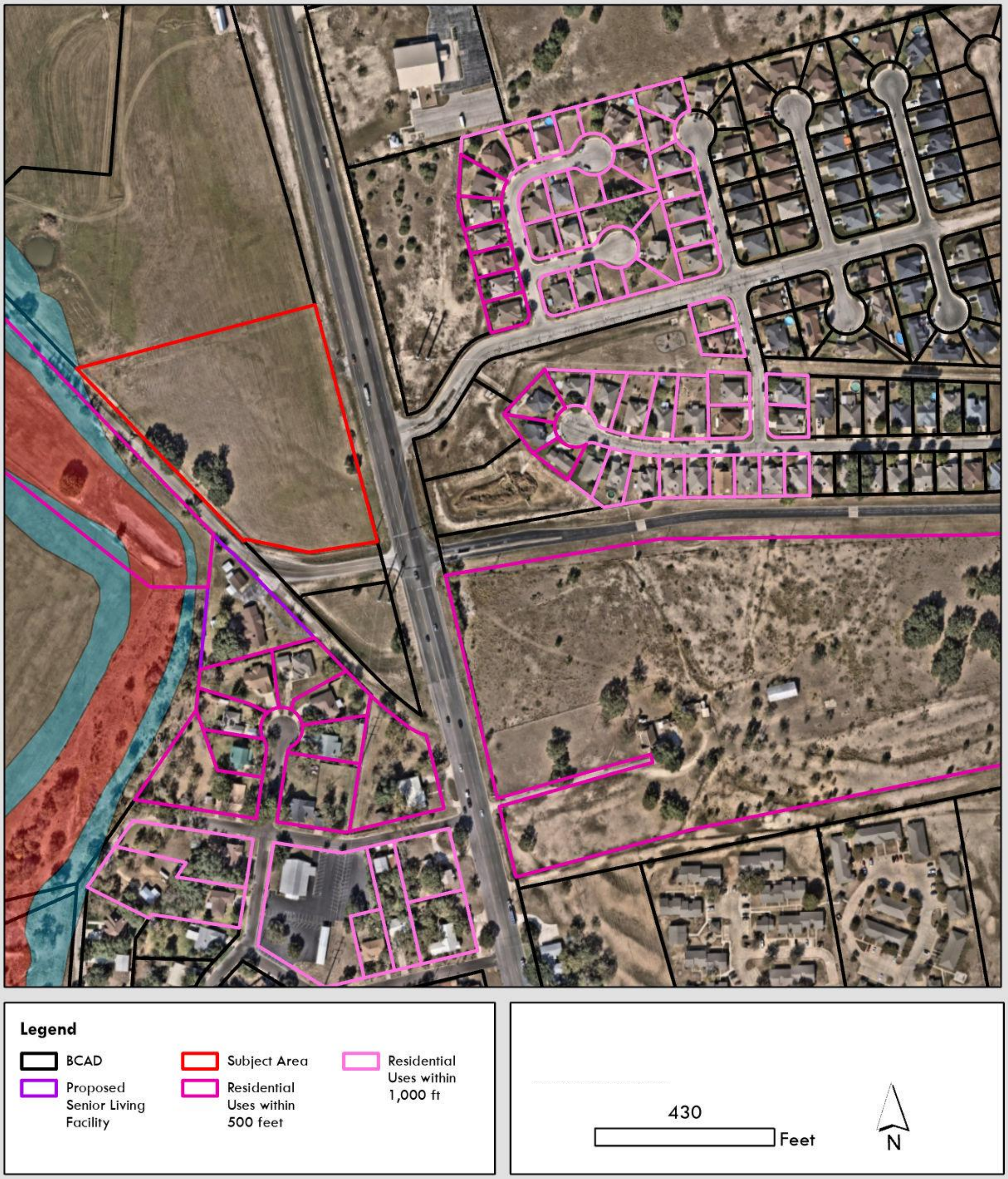
Additionally, a **2021 study** titled *Benzene emissions from gas station clusters: a new framework for estimating lifetime cancer risk* (PubMed ID: 34150235) shows increased cancer risks in communities with multiple nearby gas stations. This same study reinforces the need for a **minimum 500-foot buffer** between gas stations and places where people spend extended periods of time, such as their residence.

Furthermore, **a 2020 study by Dr. Markus Hilpert** demonstrated that 88% of vehicles still release gasoline vapors during refueling, even with Onboard Refueling Vapor Recovery systems. This suggests that current technologies are not sufficient to fully protect the public from harmful emissions.

Visual Evidence:

To illustrate the proximity of the proposed truck stop to sensitive areas, please refer to the following map:

Location of Proposed Truck Stop and Residential Areas



Note: This map is for illustrative purposes. For detailed property boundaries and measurements, please consult the Burnet Central Appraisal District Interactive Map.

Traffic Impact and Infrastructure Concerns:

The proposed use will significantly increase **truck and vehicular traffic** on **County Road 108**, which is a narrow, rural road not designed for heavy truck volumes or high-capacity ingress/egress. This raises several serious concerns:

- A **Traffic Impact Analysis (TIA)** should be required prior to any approval to evaluate the increased strain on CR 108 and the surrounding intersections.
- The road's current condition is **not adequate** to support commercial truck stop activity without **substantial upgrades**, including widening, turn lanes, and traffic controls.
- Increased traffic would **impede access for emergency vehicles** and residents, and **create safety hazards** for nearby residential neighborhoods and the senior living facility.

Impact on Public Welfare and Property Values:

Allowing a truck stop in close proximity to residential areas and senior housing will introduce:

- **24-hour diesel traffic**, noise, light pollution, and potential groundwater risks.
- **Decreased property values** due to perceived pollution and safety concerns.
- A general **decline in the character and livability** of the neighborhood.

Conclusion:

In summary, this proposal violates both the letter and intent of the City's zoning ordinance. It poses a **clear risk to public health**, **diminishes the quality of life for current and future residents**, **lacks critical infrastructure support, is incompatible with adjacent land uses,** and **could reduce surrounding property values** due to noise, traffic, and toxic air emissions. For these reasons, I respectfully urge the City Council to deny the conditional use permit.

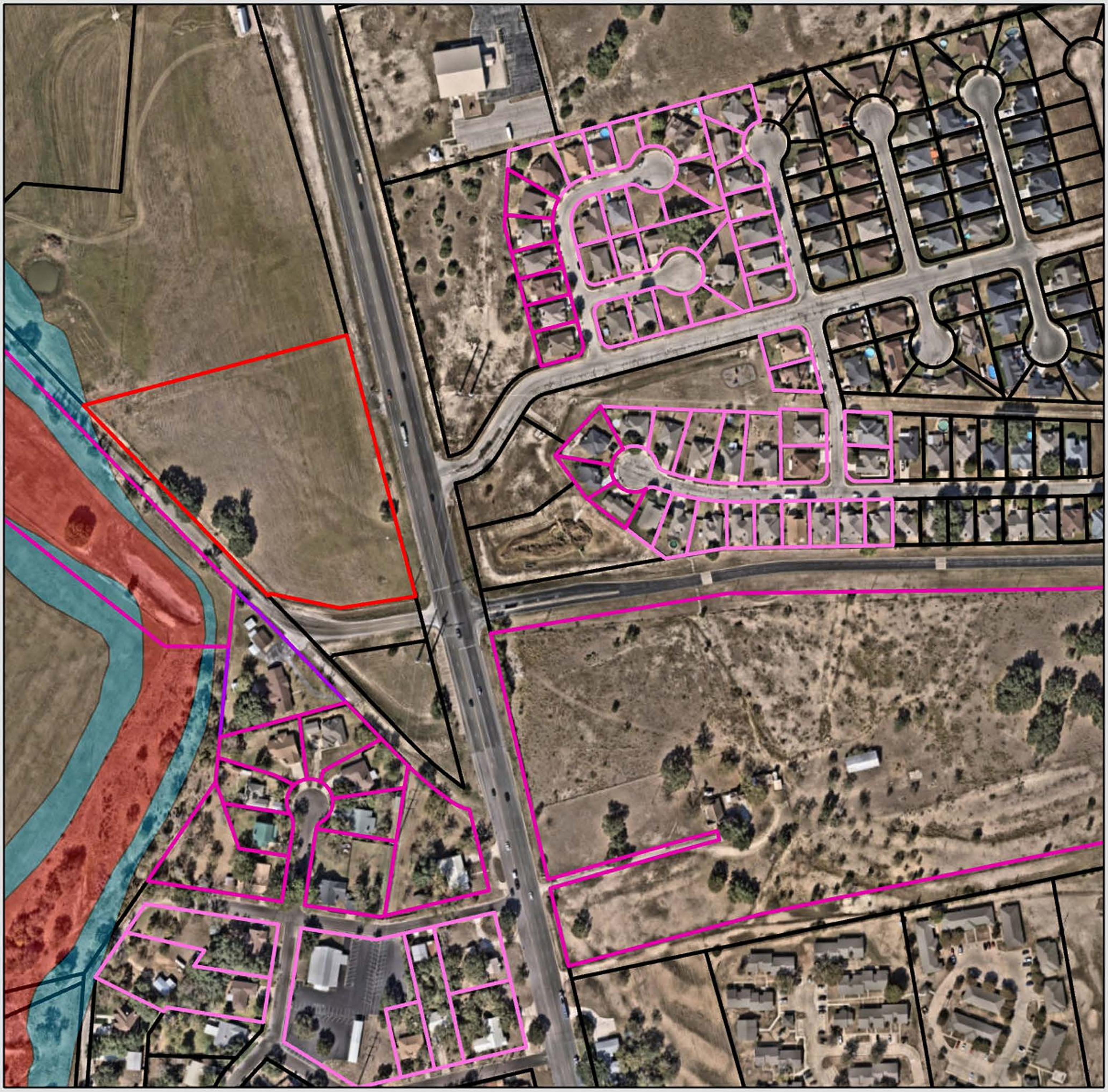
Thank you for your consideration and for your commitment to preserving the health and quality of life in Burnet.

Sincerely,
Kristen Jones

Attachments:

1. Map illustrating the proximity of the proposed truck stop to the senior living facility and surrounding residential areas.
2. Copies of the referenced studies on benzene emissions and health risks.

Location of Proposed Truck Stop and Residential Areas



Legend

- ▭ BCAD
- ▭ Proposed Senior Living Facility
- ▭ Subject Area
- ▭ Residential Uses within 500 feet
- ▭ Residential Uses within 1,000 ft

Note: This map is for illustrative purposes. For detailed property boundaries and measurements, please consult the Burnet Central Appraisal District Interactive Map.

430

Feet

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2015 Study
Hydrocarbon Release During Fuel Storage and Transfer at Gas Stations: Environmental and Health Effects

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DOI 10.1007/s40572-015-0074-8

AIR POLLUTION AND HEALTH (JD KAUFMAN AND SD ADAR, SECTION EDITORS)

Hydrocarbon Release During Fuel Storage and Transfer at Gas Stations: Environmental and Health Effects

Markus Hilpert[1] · **Bernat Adria Mora**[1] · **Jian Ni**[2] · **Ana M. Rule**[1] · **Keeve E. Nachman**[1]

Abstract At gas stations, fuel is stored and transferred between tanker trucks, storage tanks, and vehicle tanks. During both storage and transfer, a small fraction of unburned fuel is typically released to the environment unless pollution prevention technology is used. While the fraction may be small, the cumulative release can be substantial because of the large quantities of fuel sold. The cumulative release of unburned fuel is a public health concern because gas stations are widely distributed in residential areas and because fuel contains toxic and carcinogenic chemicals. We review the pathways through which gasoline is chronically released to atmospheric, aqueous, and subsurface environments, and how these releases may adversely affect human health. Adoption of suitable pollution prevention technology should not only be based on equipment and maintenance cost but also on energy- and health care-saving benefits.

Keywords Gas stations · Vapor emissions · Fuel spills · Adverse health effects · Pollution prevention

Introduction

The primary function of gas stations is to provide gasoline and diesel fuel to customers, who refill vehicle tanks and canisters. Operating a gas station requires receiving and storing a sufficient amount of fuel in storage tanks and then dispensing the fuel to customers. During delivery, storage, and dispensing of fuel at gas stations, unburned fuel can be released to the environment in either liquid or vapor form. Fuel is a complex mixture of chemicals, several of them toxic and carcinogenic [1]. Of these chemicals, the health consequences of chronic benzene exposure are best understood. Occupational studies have linked benzene exposures to numerous blood cancers, including acute myeloid leukemia and acute non-lymphocytic leukemia [2]. Concerns have been raised that gasoline vapor exposures incurred by gas station attendants [3] and tanker truck drivers [4] may result in health risks.

The potential for fuel released to the environment at gas stations, in the form of liquid spills or vapor losses, to elicit adverse health outcomes could be substantial due to the widespread distribution of gas stations across communities and the intensive usage of vehicle fuel in industrialized nations. For example, the USA consumed about 137 billion gallons of gasoline, or about 430 gallons per US citizen, in 2014 [5]. If only a small fraction of this gasoline was to be released to the environment in the form of unburned fuel, for instance 0.1 %, then about 1.6 L of gasoline would be released per capita per year in the USA. In Canada, a study estimated that evaporative losses at gas stations in 2009 amounted to 58,300,000 L [6]. With a population of about 34 million, we estimated that about 1.7 L of gasoline was released per capita per year in Canada from evaporative losses, without counting the liquid spills. While personal intake of this quantity of gasoline would result in serious adverse health effects, environmental dilution can decrease personal exposure. An overarching question is under which conditions dilution in the aqueous and atmospheric environments can limit personal exposures to acceptable levels. For example, cumulative adverse health effects could be more pronounced in metropolitan areas where more people

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This article is part of the Topical Collection on *Air Pollution and Health*

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are exposed and where the density of gas stations is larger than in rural areas.

Engineers and regulators have paid a lot of attention to leaking underground storage tanks (LUSTs) and leaky piping between storage tanks and gasoline-dispensing stations, which can result in catastrophic fuel release to the subsurface [7]. For instance, double-walled tanks have become standard in order to minimize accidental release of liquid hydrocarbon. Technologies that prevent pollution due to non-catastrophic and unreported releases of hydrocarbon that occur during fuel storage and transfer (hereafter referred to as “chronic releases”), however, have not been uniformly implemented within the developed world. The state of California in the USA has the strictest policies to minimize chronic releases, either in liquid or in vapor form. Other US states and industrialized nations, however, have not uniformly adopted California’s standards, potentially because comprehensive economic and public health analyses to inform policy making are not available. This paper focuses on chronic hydrocarbon releases at gas stations (including both liquid spills and vapor losses), their contributions to human exposures and potential health risks, and factors that influence the adoption of suitable pollution prevention technology.

Chemical Composition of Fuel

Fuels have historically contained significant fractions of harmful chemicals, some of which have been documented as contributing to morbidity and mortality in exposed persons. Crude oil, from which fuels have historically been refined, already contains toxic chemicals such as benzene [8]. Fuel additives including anti-knocking agents and oxygenates have historically also been a health concern [9]. Fuel composition has changed over time, primarily due to environmental and health concerns [9]. Fuel composition also depends on geographic location and fuel type (e.g., conventional versus reformulated gasoline) [10]. In the 1920s, lead was added to gasoline as an anti-knocking agent to replace added benzene because of its carcinogenicity [11]. Due to the massive release of lead to the environment and its neurotoxicity [12], lead was replaced in the 1970s by less toxic anti-knocking agents including methyl tert-butyl ether (MTBE) [13]. To reduce formation of ground-level ozone and associated adverse respiratory health effects [14], cleaner burning of fuel was sought in the 1990s by adding oxygenates to gasoline. This was accomplished by increasing the concentrations of MTBE, which acts as an oxygenate [9]. However, MTBE accidentally released to the subsurface [15] contaminated downstream drinking water wells relatively quickly, moving almost with the speed of groundwater, because MTBE is hydrophilic and poorly biodegradable [16]. MTBE was later on identified as a potential human carcinogen [16]. In the USA, MTBE was therefore phased out in the 1990s; at the same time, refineries began supplementing fuel with ethanol as an oxygenate [17].

In current gasoline formulations, benzene, toluene, ethylbenzene, and xylene (BTEX) and particularly benzene are the most studied chemicals and are currently believed to be of greatest health concern [18]. Table 1 shows that fuels have historically contained large fractions of toxic and carcinogenic chemicals. In many countries, lead and MTBE are no longer used. Benzene levels in gasoline are currently much lower in most countries (e.g., on average 0.62 % by volume in the USA), though the chronic health effects of benzene and other BTEX chemicals at relevant exposure levels are not well understood.

Chronic Release and Environmental Transport of Contaminants from Fuel

At gas stations, fuel can be released in both liquid and vapor phases during delivery, storage, and dispensing. Direct vapor release is usually associated with atmospheric pollution, while liquid spillage is commonly associated with soil and groundwater contamination. However, spilled liquid fuel also evaporates into the atmosphere. Hypothetically, hydrocarbon vapors can also condense back into liquid form; however, this appears to be unlikely due to quick dilution in a typically turbulent atmosphere. Figure 1 depicts how releases of unburned fuel contaminate the atmospheric, subsurface, and surface water environments (omitting LUST and leaky piping as well as marine gas stations which may release fuel directly to surface water).

Liquid Fuel Spills

Liquid fuel spills at the nozzle have received less attention than liquid releases due to LUSTs. These fuel spills occur when the dispensing nozzle is moved from the dispensing station to the vehicle tank and vice versa, when the automatic shutoff valve fails, due to spitback from the vehicle tank after the shutoff has been activated, and when the customer tops off the tank.

Table 1 Historical content of non-negligible amounts of toxic and carcinogenic chemicals in fuel

Chemical of concern	Fraction	Health effects
Benzene	Up to 5 % [75]	Carcinogenic [2]
Lead	Up to 2 g per gallon [76]	Central nervous system [12]
MTBE	Up to 15 % [77]	Potential human carcinogen [78]

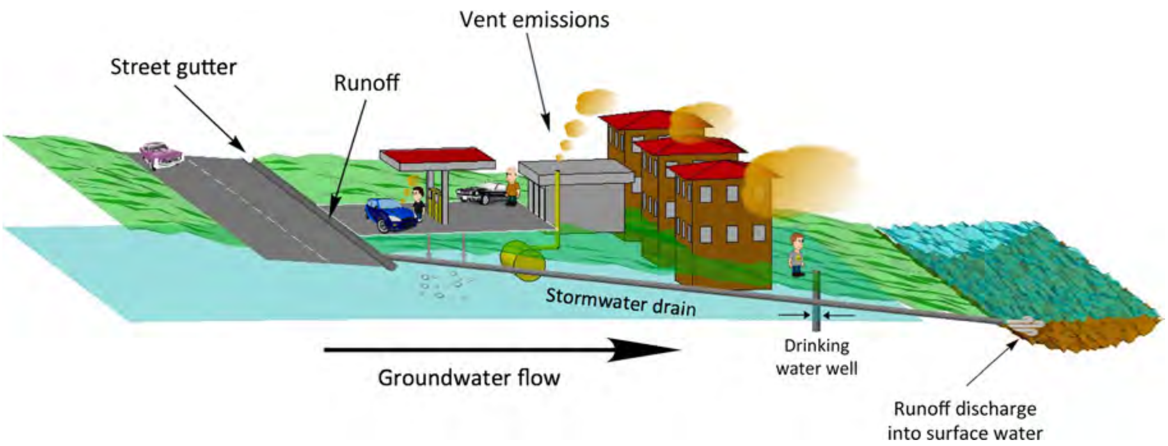

Fig. 1 Gas stations are embedded into the natural environment and can consequently release pollutants to the atmosphere, the subsurface including soil and groundwater, and surface water

In a study quantifying fuel spill frequencies and amounts at gas stations in California, about 6 L of gasoline was spilled per 16,200 gallons of gasoline dispensed at gas stations without stage II vapor recovery compared to 3.6 L at gas stations per 14,043 gallons of gasoline dispensed at gas stations with stage II vapor recovery (at the nozzle) [19]. This would mean that about 0.007 and 0.01 % of dispensed gasoline are spilled in liquid form during vehicle refueling at gas station with and without stage II recovery (numbers calculated using the assumed fuel density of 6.2 pounds/gallon). On the other hand, a study sponsored by the American Petroleum Institute found that more spills occurred at gas stations with stage II recovery [20].

We have recently performed laboratory experiments to examine the fate of liquid spill droplets. Following our previous protocol [21•], we spilled fuel droplets onto small concrete samples and measured the mass added to the concrete as a function of time. This added mass is the sum of the masses of the sessile fuel droplet and the infiltrated fuel. Figure 2 shows results for diesel and gasoline. After a certain period of time, the sessile droplet vanishes and the measured mass levels off. The remaining mass represents the infiltrated portion. The evaporated mass can be obtained by subtracting the infiltrated mass from the initial droplet mass m_0. Evaporation is greater for gasoline, while infiltration is greater for diesel spills. This is because gasoline is more volatile than diesel. Diesel has therefore a higher potential for soil contamination because of the higher infiltrated mass.

Spilled fuel may move downward in liquid or vapor phase and potentially reach the groundwater table. The physical mechanisms that govern subsurface movement of spilled fuel are the same as for fuel released due to LUST, except that spilled fuel must first penetrate relatively impermeable pavement underneath fuel-dispensing stations. Gasoline and diesel will not penetrate the groundwater table as a liquid, because they have densities lower than that of water. Released fuel may also evaporate within the sediment, and a portion of it will move downward as a vapor and potentially reach the groundwater table [22]. Whether the fuel reaches groundwater in liquid or vapor form, the fuel will then partition into groundwater and become a dissolved chemical that is carried away by molecular diffusion and groundwater flow and associated hydrodynamic dispersion [23]. Therefore, the spills can contaminate downstream drinking water wells [24]. Biodegradation can decrease contaminant concentrations significantly; however, its efficiency depends on many factors including the chemical composition of the fuel and the presence of suitable microbial species that can metabolize a given contaminant, bioavailability, and electron acceptor availability [25]. Partitioning of the contaminant into other phases will cause

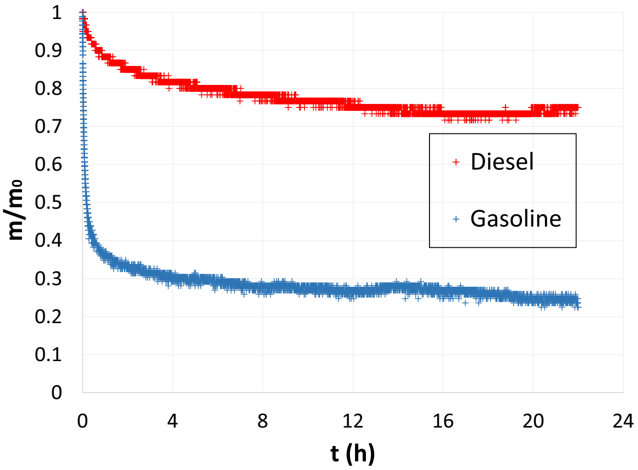

Fig. 2 Results from laboratory experiments, in which we spilled a mass m_0=1 g of diesel or gasoline onto concrete samples. The measured mass m represents the masses of the sessile droplet and infiltrated liquid

retarded transport of the contaminant within groundwater. For instance, hydrophobic contaminants such as benzene tend to sorb to the sediment. For this reason, large-scale contamination of aquifers and associated adverse health effects due to the ingestion of contaminated drinking water from these aquifers are often considered a lesser concern for hydrophobic contaminants [16].

Stocking et al. [26] evaluated the potential of groundwater contamination due to small one-time releases of liquid gasoline. In a case study, they assumed a spill volume much bigger than the ones typically measured by the study of gas stations in California [19], i.e., 0.5 L, and they concluded the risk to groundwater to be small. This analysis, however, did not include consideration of a key mechanism for fuel spillage; namely, that much smaller droplets are typically released during vehicle refueling [19]. To address this question, Hilpert and Breysse [21•] calculated cumulative spill volumes due to repeated small spillages that occur at gasoline-dispensing facilities and estimated that a gas station selling about 400,000 L of gasoline per month would spill at least 150 L each year. They also developed a model that shows that the fraction of spilled gasoline that infiltrates into the pavement increases as the droplet size decreases. Therefore, repeated small spills could be of greater concern for groundwater contamination than an instantaneous release of the cumulative spill volume; thus, a risk to groundwater may not be as small as previously estimated.

Laboratory experiments and modeling have shown that gasoline from small-volume spills can infiltrate into the concrete that usually covers the ground underneath gasoline-dispensing stations—despite the low permeability of concrete and the high vapor pressure of gasoline [21•]. It is unlikely that liquid fuel fully penetrates a concrete slab to contaminate the underlying natural subsurface due to the low permeability of concrete [27], although preferential pathways for fluid flow such as cracks and faulty joints between concrete slabs can allow for such liquid penetration. It has been hypothesized that evaporation of infiltrated gasoline and subsequent downward migration of the vapor through the concrete may lead to contamination of underlying sediment and groundwater [21•]. Consistent with these two proposed pathways of subsurface contamination, soil/sediment underneath concrete pads of a gas station in Maryland was contaminated by diesel oil and gasoline (leaky piping could have also contributed to the contamination) [28].

Runoff water that flows over pavement can also get contaminated with hydrocarbons spilled onto the pavement [29–31], and such contamination has specifically been linked to gas stations [32–34]. If a spill occurs while runoff occurs, the hydrocarbon can be expected to float on top of the water sheet, because gasoline, diesel oil, and lubricants are typically less dense than water (light non-aqueous phase liquids or LNAPLs). While runoff water is not directly ingested, it is funneled into the stormwater drainage system, and may be released to natural water bodies, often without treatment. Whereas volatilization decreases contaminant levels in the stormwater within hours depending on the exact environmental conditions [35], and biodegradation will further decrease levels, significantly contaminated stormwater might be released to natural water bodies if they are close by. Finally, fuel spilled at marine gas stations may directly enter natural water bodies.

Vapor Fuel Releases

Fuel evaporative losses have received more attention than liquid fuel spills (even though they are related) [6, 36]. These losses are related to the fact that the headspace above liquid fuel in vehicle and storage tanks tends to approach thermodynamic equilibrium with the liquid. Consequently, almost saturated gasoline vapors can be released to the atmosphere when tanks are refueled, unless a suitable vapor recovery system is in place. Since saturated gasoline vapors have a density that is three to four times larger than the one of air, i.e., 4 kg/m^3, and the density of liquid gasoline is about 720 kg/m^3 [37], about 0.5 % of liquid gasoline dispensed to a tank is released to the atmosphere if the entire headspace is in equilibrium with the liquid fuel. This is true for any type of tank, whether it is a vehicle tank, a canister, an underground storage tank (UST), or an above-storage tank. The percentage loss is less if a tank received clean air relatively recently, e.g., when the fuel level in a storage tank drops because of gasoline-fuel dispensing.

It is important to note that vapor recovery at the nozzle can cause vapor releases at the storage tank, because vapors recovered at the nozzle are typically directed into the storage tank. The storage tank, in turn, can “breathe” and potentially release recovered vapors immediately or at a later time. A tank sucks in relatively uncontaminated air as the liquid fuel level drops in the tank due to vehicle refueling, and it releases vapors through the vent pipe into the atmosphere if the gas pressure increases and exceeds the cracking pressure of the pressure/vacuum valve, when fuel evaporates into unequilibrated gas in the headspace.

As discussed in the “Liquid Fuel Spills” section above, we note that liquid spills also contribute to air pollution because spilled droplets form sessile droplets on pavement that can then evaporate into the atmosphere. On concrete, most of spilled gasoline droplets evaporate into the atmosphere (Fig. 2). This, however, does not mean that the small fraction that infiltrates into the concrete is not of concern.

Exposure and Risks to Human Populations

Gas stations exist as part of the built environment and are widely distributed across communities. As a result, they may be surrounded by residential dwellings, businesses, and other buildings such as schools. Operation of gas stations may thus create opportunities for a variety of human populations to be exposed to vapors during station tank filling and vehicle refueling. These human populations can be broadly grouped into three groups: populations exposed occupationally as a result of employment in various capacities at the service station; those exposed as customers engaging in vehicle refueling; and those passively exposed either by residing, attending school, or working near the refueling station. The exposures to benzene and other components of refueling vapors and spills experienced by these populations vary based on a number of factors, including the size and capacity of the refueling station, spatial variation in pollutant concentrations in ambient air, climate, meteorological conditions, time spent at varying locations of the service station, changing on-site activity patterns, physiological characteristics, and the use of vapor recovery and other pollution prevention technologies.

Employees at service stations (such as pump attendants, on-site mechanics, and garage workers) are among those with greatest exposure to benzene originating from gas stations [3]. These receptors spend the most time on site (potentially reflecting approximately 40 h per week, for decades) and intermittently spend time where vapors from the pump are at their highest concentrations, with benzene concentrations measuring between 30 and 230 ppb in the breathing zone [38–40]. Gas station patrons can also be exposed to vapors when refueling. Compared to station employees, their exposures are brief and transient. A Finnish study reported a median time spent refueling of approximately 1 min, whereas 3 min was the median duration in the USA [41, 42]. The same US study reported an average benzene personal exposure concentration at the pump of 910 ppb, with the strongest predictors of benzene levels being fuel octane grade, duration of exposure, and season [42].

Those occupying residences, businesses, and other structures neighboring gas stations can also be exposed to fuel vapors originating in the gas station, though typically at lower concentrations than those measured at the pump. While vapor concentrations will drop as the distance from the service station increases, exhaust fumes from waiting customers and fuel delivery trucks can also contribute to vapors in proximity to gas stations. A small number of studies have examined benzene concentrations at the fenceline of the service station and beyond. A study published by the Canadian petroleum industry found average benzene concentrations of 146 and 461 ppb at the gas station property boundary in summer and winter, respectively [43]. A South Korean study examined outdoor and indoor benzene concentrations at numerous residences within 30 m and between 60 and 100 m of gas stations and found median outdoor benzene concentrations of 9.9 and 6.0 μg/m^3 (about 3.1 and 1.9 ppb), respectively. Median indoor concentrations at these locations were higher, reaching 13.1 and 16.5 μg/m^3 (about 4.1 and 5.2 ppb), respectively [44]. Another study found median ambient benzene levels of 1.9 ppb in houses both <50 and >100 m from a service station [45]. Yet, another study [46] found that benzene and other gasoline vapor releases from service stations can be discerned from traffic emissions as far as 75 m from service stations and that the contribution of service stations to ambient benzene is less important in areas of high traffic density. This is because vehicle exhaust is usually the most abundant volatile organic compound (VOC) in urban areas, often followed by gasoline vapor emissions from fuel handling and vehicle operation [47].

Beyond contact with surface-level gasoline vapors, fuel releases may result in other exposure pathways. Soil and groundwater contamination is common at gas stations. Drinking water wells proximate to gas stations, which in rural areas are often the only drinking water source, can become contaminated, potentially exposing well users to benzene and other chemicals [48, 49]. In addition, runoff from rain and other weather events can carry spilled hydrocarbons, which can contaminate surface waters; those using surface waters, either recreationally or for other purposes, may be exposed to these contaminants through dermal contact or incidental ingestion.

In the USA, the Environmental Protection Agency (EPA) regulates releases of benzene under the Clean Air Act as a hazardous air pollutant, and benzene is listed as number 6 on the 2005 priority list of hazardous substances under the Comprehensive Environmental Response, Compensation, and Liability Act and any release greater than 10 pounds triggers a reporting requirement. Different quantitative toxicity metrics exist for benzene inhalation. The EPA Integrated Risk Information System (IRIS) has published a reference concentration of 0.03 mg/m^3 (about 9.4 ppb), corresponding to decreased lymphocyte counts [50], whereas the NIOSH recommended exposure limit (REL) is a time-weighted average concentration (for up to a 10-hour workday during a 40-hour workweek) of 0.319 mg/m^3 (about 100 ppb) [51].

While research attention has been paid to measurement of gasoline vapor constituent concentrations in air at and near service stations, less is known about the health consequences faced by those that are exposed to gasoline vapors. Of the limited literature examining these exposures, service station workers have received the greatest attention, and exposure is often assessed as a function of job title, rather than specific measurements of vapor constituent concentrations. An older study looking broadly at leukemia incidence in Portland, Oregon, found that gas station workers were at significantly increased risk for lymphocytic leukemia [52]. A proportionate mortality ratio analysis of all deaths recorded in New Hampshire among white men from 1975 to 1985 found elevated leukemia mortality in service station workers and auto mechanics [53]. The type of leukemia was not specified. An Italian occupational cohort study of refilling attendants that examined risks among workers at smaller gas stations reported non-significant increases in mortality for non-Hodgkin's lymphoma and significantly elevated mortality for esophageal cancer in men, as well as increased brain cancer mortality in both sexes [54]. A different cohort of 19,000 service station workers in Denmark, Norway, Sweden, and Finland examined an array of cancer end points and found increased incidence for multiple sites (nasal, kidney, pharyngeal, laryngeal, and lung) among workers estimated to be occupationally exposed to benzene in the range of 0.5–1 μg/m^3 (0.16 - 0.31 ppb). Non-significant increased incidence was found for acute myeloid leukemia in men and for leukemia different from acute myeloid leukemia and chronic lymphocytic leukemia in women [55]. A case–control study of multiple occupations including subjects from the USA and Canada found significant increases in rates of total leukemia and acute myeloid leukemia but not acute lymphocytic leukemia in gas station attendants [56]. A 2015 review of studies examining potential relationships between benzene exposures and hematopoietic and lymphatic cancers among vehicle mechanics yielded inconclusive results, although it suggested that if an effect was to exist, it would be small and difficult to rigorously ascertain with existing epidemiologic methods [57].

The health consequences of nearby residents of gas stations have not been studied. However, it is known that contaminated groundwater can affect large numbers of people if the groundwater is used as drinking water, as was the case in Camp Lejeune (North Carolina, USA) where thousands were exposed to a range of chemicals including gasoline released from LUSTs [58]. A study of Pennsylvania residents residing in close proximity to a large gasoline spill from a LUST found evidence of increased leukemia risks [49, 59••]. The health consequences of chronic fuel releases at gas stations that can, for example, occur due to ingestion of contaminated groundwater, fuel vapor intrusion from contaminated soil and groundwater into dwellings [60], and atmospheric vapor releases during fuel transfer and storage have not been studied. While limited measurements of ambient concentrations of vapor constituents in communities were identified, literature searches did not identify studies of the health consequences of inhalation exposures to gasoline vapors among community residents [61].

Pollution Prevention

Pollution prevention technologies have been developed that can efficiently reduce the releases of unburned fuel to the environment that routinely occur during fuel storage and transfer (see Fig. 3):

1. Stage I vapor recovery collects vapors that would be expelled from USTs during fuel delivery [62]. Without stage I vapor recovery, about 80 kg of gasoline vapor would be released from a 40 m^3 UST if one assumes a saturated vapor density of 4 kg/m^3 [37] and vapors in the headspace

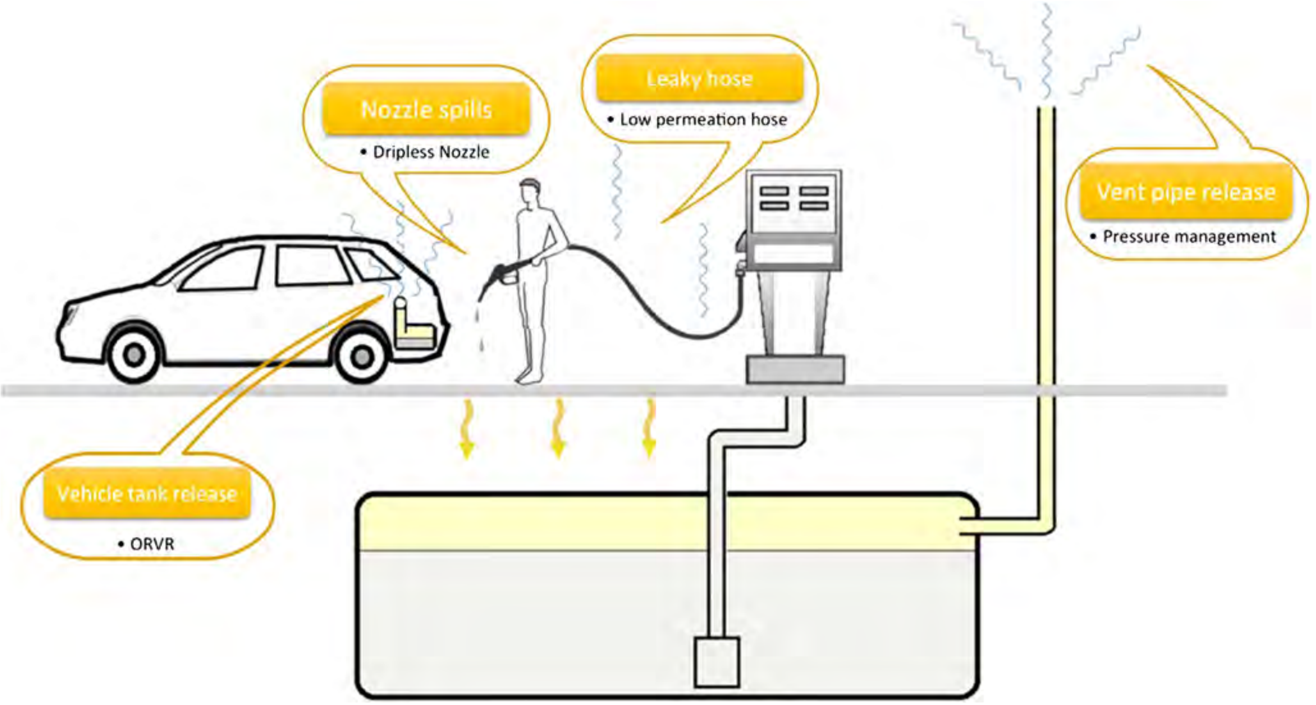

Fig. 3 There are several sources of chronic release of unburned fuel at gas stations that occur due to fuel storage and dispensing: vapor release through the vent pipe of the storage tank, vapor release from the vehicle tank during refueling, leaky dispensing hoses, liquid spills during vehicle refueling, and vapor emissions through evaporation of this spilled fuel. As indicated, suitable pollution prevention technology can minimize the releases. Onboard refueling vapor recovery (ORVR)

to be at half saturation. Stage I vapor recovery can thus prevent substantial fuel vapor releases that would occur within a short period of time. Such releases might expose tanker truck drivers and persons in the proximity of a gas station to significant doses of fuel vapors. Stage I vapor recovery is accomplished by establishing a closed loop between the UST and the tanker truck. Through a fuel delivery hose, liquid fuel is pumped into the UST, while a vapor recovery hose directs vapors displaced from the UST into the headspace of the tanker truck. Stage I vapor recovery is currently required for high-throughput gas stations in all states in the USA and in most countries.

2. Stage II vapor recovery technology can efficiently collect vapors expelled from vehicle tanks during refueling, thereby minimizing personal exposure of customers and workers to fuel vapors during dispensing of gas [63]. Recovered vapors are directed into the UST. Two technologies for stage II vapor recovery have been developed, the vaccum-assist method and the balance method. In the vacuum-assist method, contaminant-laden air is actively removed/pumped from the nozzle into the UST. In the balance method, displaced vapors are passively withdrawn by connecting the vapor recovery hose to the inlet of the vehicle tank via an airtight seal. The pressure increase in the headspace of the vehicle tank provides a driving force that seeks to push the vapors into the storage tank. Stage II vapor recovery has been required in many states of the USA and in other countries, although there is currently an effort to decommission stage II vapor recovery (see below).
3. Technology development at the hose and nozzle level can also contribute to reduced fuel releases. Low-permeation hoses, for instance, limit the release of gasoline vapors through the wall of the refueling hoses [64]. Dripless nozzles have been developed to minimize liquid spills that occur when the nozzle is moved between the fill pipe and the dispensing unit.
4. Passenger vehicles and trucks can be equipped with onboard refueling vapor recovery (ORVR) systems which direct vapors that, during vehicle refueling, would be released to the atmosphere into an activated carbon-filled canister in the vehicle [65, 66]. Collected vapors are later reintroduced into the vehicle's fuel system. However, canisters, motorcycles, and boats are not equipped with ORVR.
5. Impermeable liners underneath the concrete pads can reduce the risk of soil and groundwater contamination once environmental fuel releases, in liquid or vapor phase, have occurred. However, this technology might eventually result in air pollution, because liquid fuel that is hindered from moving downward in the concrete pad will tend to saturate the pavement and eventually evaporate into the atmosphere.
6. Finally, unburned fuel vapor can be released from an UST when the tank pressure exceeds the cracking pressure of the pressure/vacuum valve and it can be prevented by two pressure management techniques, burning or separation of air and fuel vapors. Released air/fuel vapors can be burned, however, which results in the release of combustion-related pollutants into the atmosphere. Alternatively, a semi-permeable membrane can be used to separate the air from the fuel vapors. Depressurization of the tank is then achieved by releasing the relatively clean air through the pressure/vacuum valve to the atmosphere.

When it comes to evaluating the efficiency of vapor recovery during liquid transfer between tanks, it is of upmost importance to consider potential releases from all tanks; they form a system. Otherwise, the overall efficiency of stage II vapor recovery cannot be understood. For instance, stage II vapor recovery based on the vacuum-assist method can negatively interfere with ORVR. In that case, no vapors are released from the vehicle tank and the stage II pump draws relatively clean air from the atmosphere into the storage tank. In the UST, this air will become saturated with fuel vapors that evaporate from the stored fuel. This results in pressurization of the UST and release of contaminant-laden air if the tank pressure exceeds the cracking pressure of the pressure/vacuum valve of the UST. This might occur immediately or at a later point in time. However, there are stage II systems that do not negatively interfere with ORVR including the balance method.

Estimates for the efficiency of pollution technologies are usually provided by the manufacturers. However, adoption of these technologies by gas station owners usually relies on the certification and quantification of efficiencies by independent parties. In the USA, the California Air Resources Board and EPA typically assume this role [36]. Consultants and environmental agencies have used these estimates to determine current releases of unburned fuel to the environment and to evaluate the effects of pollution prevention technology [67].

While many studies have found health benefits from pollution prevention technology intended to minimize chronic gasoline spills, these studies typically do not quantify overall financial benefits and costs. Instead, only equipment and maintenance cost are typically considered [68]. Adopting the new equipment can reduce fuel losses and reduce environmental cost and health risks. However, this new equipment comes with non-trivial upfront costs. It is therefore a concern that the related policy-making process of chronic fuel spills relies only on non-comprehensive cost estimates. Studies are needed that account for health care cost due to released pollutants and energy-saving benefits due to pollution prevention. Such econometric studies have, for example, been performed in the context of pollutant emissions from coal-fired power plant and commercial real estate development [69••, 70]. At times, there is also the perception that pollution prevention costs are only carried by the specific industry [71]. Adoption of the environmentally friendly technology could be slow when the firms have long equipment replacement cycles or when the firms do not have sufficient information to evaluate whether or not a switch to an environmentally friendly technology is in their private interests. It is, however, not clear that this apparent investment, in the form of prevention cost, might also be partly shouldered by customers and that this apparent cost might actually (at least in the long run) be beneficial to customers, gas station workers, nearby residents, and other populations that spend significant amounts of times in the proximity of gas stations (e.g., school children in nearby schools). Policy intervention is often expected to expedite the adoption of such environmental friendly technologies, in order to reduce the difference in the private and social values of adoption.

Efforts are currently underway that could potentially allow decommissioning stage II vapor recovery in the USA due to the widespread use of ORVR in the motor vehicle fleet [68]. However, the remaining legacy fleet without ORVR and all motorcycles and boats (lacking ORVR) can produce significant emissions during vehicle refueling, emissions that could be avoided by stage II vapor recovery. For the State of Maryland, it has been estimated that fuel consumption of non-ORVR-equipped vehicles was about 10 % in 2015 (Table 4 in [67]). These emissions can result in direct hydrocarbon exposures among vehicle owners during vehicle refueling as well as in passive exposure of other populations. A comprehensive cost analysis of the decommissioning of stage II recovery represents an opportunity to inform policy makers on their recommendation with regards to stage II recovery.

Conclusions

Even if only a small fraction of unburned fuel is lost during vehicle refueling and fuel storage, the cumulative release of fuel to the environment can be large if large total amounts of fuel are dispensed at gas stations. For instance, about 0.01 % of fuel can be spilled during the refueling process and up to about 0.5 % can be lost in vapor form if equilibrated gasoline vapors are released from a tank to the atmosphere during refueling (worst-case scenario). For a medium-size gas station, which sells 400,000 L of gasoline per month, this results in 480 L of spilled gasoline and in 24,000 L of liquid gasoline that is anually released in vapor form to the environment. Even though dilution can reduce concentrations of released contamination, research is needed to assess whether such releases represent an environmental health concern.

The potential for pollution prevention, moreover, is substantial. Technology has already been developed and partially employed that can efficiently decrease vapor losses and liquid spills. Particularly, when it comes to vapor losses, it is crucial to consider not only vapor recovery at the vehicle tank/nozzle but also at the storage tank, since vapors recovered at the nozzle are directed into the storage tank, from which they might be potentially released. While California has implemented the strictest regulations when it comes to preventing chronic hydrocarbon releases at gas stations, other highly industrialized states and nations do not employ the same standards for different reasons. For instance, pressure/vacuum valves on vent pipes of fuel storage tanks are not common in Canada, because they might freeze in the wintertime, potentially causing a tank implosion [6].

Relatively little research has been done on potential soil and groundwater contamination due to chronic releases of liquid fuel during vehicle refueling. Unlike catastrophic releases, such as LUST, chronic spills are not reported. Limited field investigations suggest that spilled fuel may penetrate concrete underneath dispensing pads to contaminate underlying sediment. However, it is possible that such soil contamination occurs routinely over the life span of a gas station and that this contamination pathway is masked or erroneously explained by leaks in the piping from the USTs to the dispensers. Overall, large-scale soil and groundwater contamination by fuel appears to be a lesser problem, because many of the toxic compounds in fuel are hydrophobic (including BTEX) and can therefore be expected not to travel too far in groundwater. However, customers, gas station workers, and nearby residents may get exposed to the hydrocarbons if groundwater is used as a drinking water supply or if fuel vapor intrusion in dwellings occurs.

Health effects of living near gas stations are not well understood. Adverse health impacts may be expected to be higher in metropolitan areas that are densely populated. Particularly affected are residents nearby gas stations who spend significant amounts of time at home as compared to those who leave their home for work because of the longer period of exposure. Similarly affected are individuals who spend time close to a gas station, e.g., in close by businesses or in the gas station itself. Of particular concern are children who, for example, live nearby, play nearby, or attend nearby schools, because children are more vulnerable to hydrocarbon exposure [72].

Potential future changes in fuel composition might pose new environmental health challenges as there is a history of adding even large amounts of toxic substances to fuel (Table 1). Changes in fuel composition could occur due to an increasing usage of biofuels, or to comply with air quality standards, which might also change over time. Chemicals newly added to fuel or changes in chemical concentrations can have unforeseen ramifications. One could argue that future fuel composition changes will be performed with more care; however, it was only in the 1990s, decades after the Safe Drinking Water Act (SDWA) was passed in 1974, that MTBE was added to gasoline without critically evaluating its transport behavior in groundwater and toxicity, a mistake which nowadays is considered avoidable [73]. Interestingly, ethanol, which has largely replaced MTBE, can inhibit biodegradation of BTEX, which is not the case for MTBE [74]. Given the complexities of chemical fate and transport in the environment and the potential for insufficient toxicity testing, using appropriate pollution prevention technology that minimizes release of unburned chemicals with known and unknown adverse health effects during fuel storage and transfer seems a wise, long-term, and cost effective idea given ever-changing fuel compositions.

Finally, employing efficient pollution prevention technology might be economically advantageous. The evaluation of economic benefits of pollution prevention technology needs to account not only for the cost of implementation and maintenance of such technology but also for public health burdens due to released pollutants and energy-saving benefits due to valuable hydrocarbons not wastefully released to the environment.

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Compliance with Ethics Guidelines

Conflict of Interest Markus Hilpert, Bernat Adria Mora, Jian Ni, Ana Rule, and Keeve Nachman declare that they have no conflict of interest.

Human and Animal Rights and Informed Consent This article does not contain any studies with human or animal subjects performed by any of the authors.

References

Papers of particular interest, published recently, have been highlighted as:
- • Of importance
- •• Of major importance

1. Wang Z et al. Characteristics of spilled oils, fuels, and petroleum products: 1. composition and properties of selected oils. United States Environmental Protection Agency. Report No.: EPA/600/R-03/072, 2003.
2. IARC, IARC monographs on the evaluation of carcinogenic risks to humans. Vol. 100F. 2012.
3. Karakitsios SP et al. Assessment and prediction of exposure to benzene of filling station employees. Atmospheric Environment. 2007;41(40):9555–69.
4. Javelaud B et al. Benzene exposure in car mechanics and road tanker drivers. International Archives of Occupational and Environmental Health. 1998;71(4):277–83.
5. U.S. Energy Information Administration. How much gasoline does the United States consume? 2015 [cited 2015 July 20]; Available from: http://www.eia.gov/tools/faqs/faq.cfm?id=23&t=10.
6. Statistics Canada. Gasoline evaporative losses from retail gasoline outlets across Canada 2009. Environment Accounts and Statistics Analytical and Technical Paper Series, 2012.
7. Dowd RM. Leaking underground-storage tanks. Environmental Science & Technology. 1984;18(10):A309–9.
8. Centers for Disease Control and Prevention. Facts about benzene. 2015 Available from: http://www.bt.cdc.gov/agent/benzene/basics/facts.asp.
9. Nadim F et al. United States experience with gasoline additives. Energy Policy. 2001;29(1):1–5.
10. Weaver JW , Exum LR , Prieto LM. Gasoline composition regulations affecting LUST sites. U.S. Environmental Protection Agency Office of Research and Development Washington, DC 20460, 2010. Report No.: EPA 600/R-10/001.
11. Needleman HL. The removal of lead from gasoline: historical and personal reflections. Environmental Research. 2000;84(1):20–35.
12. Grandjean P, Landrigan PJ. Developmental neurotoxicity of industrial chemicals. Lancet. 2006;368(9553):2167–78.
13. Thomas VM. The elimination of lead in gasoline. Annual Review of Energy and the Environment. 1995;20:301–24.
14. U.S. Environmental Protection Agency. National Ambient Air Quality Standards for ozone; final rule. Fed Reg. 2008;73:16436–514.
15. U.S. Environmental Protection Agency, Achieving clean air and clean water: The report of the blue ribbon panel on oxygenates in gasoline, 1999.
16. Squillace PJ et al. Review of the environmental behavior and fate of methyl tert-butyl ether. Environmental Toxicology and Chemistry. 1997;16(9):1836–44.
17. U.S. Energy Information Administration, Eliminating MTBE in gasoline in 2006. 2006.
18. U.S. Environmental Protection Agency, Fuel oxygenates (MTBE, TBA, and ethanol). 2015.
19. Morgester JJ, Fricker RL, Jordan GH. Comparison of spill frequencies and amounts at vapor recovery and conventional service stations in California. Journal of the Air & Waste Management Association. 1992;42(3):284–9.
20. Mueller EA. A survey and analysis of liquid gasoline released to the environment during vehicle refueling at service stations. Washington, DC: American Petroleum Institute; 1989.
21. • Hilpert M, Breysse PN. Infiltration and evaporation of small hydrocarbon spills at gas stations. Journal of Contaminant Hydrology. 2014;170:39–52. **This study examines for the first time the fate and transport of small fuel droplets spilled during vehicle refueling**.
22. Dakhel N et al. Small-volume releases of gasoline in the vadose zone: impact of the additives MTBE and ethanol on groundwater quality. Environmental Science & Technology. 2003;37(10):2127–33.
23. Charbeneau R.J., Groundwater hydraulics and pollutant transport 2006: Waveland Press, Inc.
24. Grady, S. and G. Casey, Occurrence and distribution of methyl tert-butyl ether and other volatile organic compounds in drinking water in the northeast and mid-Atlantic regions of the United States, 1993–98. Water Resources Investigations Report WRIR 00–4228. Geological Survey, U.S., 2001.
25. Leahy JG, Colwell RR. Microbial-degradation of hydrocarbons in the environment. Microbiological Reviews. 1990;54(3):305–15.
26. Stocking, A.S., et al., Evaluation of fate and transport of methyl tertiary butyl ether (MTBE) in gasoline following a small spill. In: Stanley, Anita, (eds.) Petroleum hydrocarbons and organic chemicals in ground water—prevention, detection, and remediation, Houston, Tex., Nov. 17–19, 1999. Proceedings.: National Ground Water Association, and American Petroleum Institute, 1999: p. 229–246.
27. Jacobs P. Permeabilität und Porengefüge Zementgebundener Werkstoffe. ETH Zürich: Switzerland; 1994.
28. Aria Environmental Inc., Underground Storage Tank Closure Report Chesapeake House – Exxon Facility (Northern Service

Station) I-95 Travel Plaza, North East, Cecil County, Maryland, 2014, Maryland Transportation Authority.

29. Latimer JS et al. Sources of petroleum-hydrocarbons in urban runoff. Water Air and Soil Pollution. 1990;52(1–2):1–21.
30. Ohe T, Watanabe T, Wakabayashi K. Mutagens in surface waters: a review. Mutation Research-Reviews in Mutation Research. 2004;567(2–3):109–49.
31. Hoffman EJ et al. Urban runoff as a source of polycyclic aromatic-hydrocarbons to coastal waters. Environmental Science & Technology. 1984;18(8):580–7.
32. Borden RC, Black DC, McBlief KV. MTBE and aromatic hydrocarbons in North Carolina stormwater runoff. Environmental Pollution. 2002;118(1):141–52.
33. Garcia MR et al. Assessment of polycyclic aromatic hydrocarbon influx and sediment contamination in an urbanized estuary. Environ Monit Assess. 2010;168(1–4):269–76.
34. Khan E, Virojnagud W, Ratpukdi T. Use of biomass sorbents for oil removal from gas station runoff. Chemosphere. 2004;57(7):681–9.
35. U.S. Environmental Protection Agency. Technical factsheet on: Benzene. 2015 Available from: http://www.epa.gov/ogwdw/pdfs/factsheets/voc/tech/benzene.pdf.
36. CARB. Vapor Recovery Program. 2015 Available from: http://www.arb.ca.gov/vapor/vapor.htm.
37. International Chemical Safety Cards (ICSC). ICSC #: 1400. 2015 Available from: http://www.cdc.gov/niosh/ipcsneng/neng1400.html.
38. van Wijngaarden E, Stewart PA. Critical literature review of determinants and levels of occupational benzene exposure for United States community-based case–control studies. Applied Occupational and Environmental Hygiene. 2003;18(9):678–93.
39. Hartle R. Exposure to methyl tert-butyl ether and benzene among service station attendants and operators. Environmental Health Perspectives. 1993;101 Suppl 6:23–6.
40. Periago JF, Zambudio A, Prado C. Evaluation of environmental levels of aromatic hydrocarbons in gasoline service stations by gas chromatography. Journal of Chromatography A. 1997;778(1–2):263–8.
41. Vainiotalo S et al. Customer exposure to MTBE, TAME, C6 alkyl methyl ethers, and benzene during gasoline refueling. Environmental Health Perspectives. 1999;107(2):133–40.
42. Egeghy PP, Tornero-Velez R, Rappaport SM. Environmental and biological monitoring of benzene during self-service automobile refueling. Environmental Health Perspectives. 2000;108(12):1195–202.
43. Akland GG. Exposure of the general population to gasoline. Environmental Health Perspectives. 1993;101 Suppl 6:27–32.
44. Jo W-K, Moon K-C. Housewives' exposure to volatile organic compounds relative to proximity to roadside service stations. Atmospheric Environment. 1999;33(18):2921–8.
45. Jo W-K, Oh J-W. Exposure to methyl tertiary butyl ether and benzene in close proximity to service stations. Journal of the Air & Waste Management Association. 2001;51(8):1122–8.
46. Terrés IMM et al. Assessing the impact of petrol stations on their immediate surroundings. Journal of Environmental Management. 2010;91(12):2754–62.
47. Watson JG, Chow JC, Fujita EM. Review of volatile organic compound source apportionment by chemical mass balance. Atmospheric Environment. 2001;35(9):1567–84.
48. Wallace LA. The exposure of the general population to benzene. Cell Biology and Toxicology. 1989;5(3):297–314.
49. Patel AS et al. Risk of cancer as a result of community exposure to gasoline vapors. Archives of Environmental Health. 2004;59(10):497–503.
50. U.S. Environmental Protection Agency. Benzene (CASRN 71-43-2). 2015 Available from: http://www.epa.gov/iris/subst/0276.htm.
51. National Institute for Occupational Safety and Health. NIOSH Pocket Guide to Chemical Hazards: Benzene. 2015 Available from: http://www.cdc.gov/niosh/npg/npgd0049.html.
52. Morton W, Marjanovic D. Leukemia incidence by occupation in the Portland-Vancouver metropolitan area. American Journal of Industrial Medicine. 1984;6(3):185–205.
53. Schwartz E. Proportionate mortality ratio analysis of automobile mechanics and gasoline service station workers in New Hampshire. American Journal of Industrial Medicine. 1987;12(1):91–9.
54. Lagorio, S., et al. Mortality of filling station attendants. Scand Journal Work Environ Health 1994: 331–338.
55. Lynge E et al. Risk of cancer and exposure to gasoline vapors. American Journal of Epidemiology. 1997;145(5):449–58.
56. Terry PD et al. Occupation, hobbies, and acute leukemia in adults. Leukemia Research. 2005;29(10):1117–30.
57. Hotz P, Lauwerys RR. Hematopoietic and lymphatic malignancies in vehicle mechanics. Critical Reviews in Toxicology. 1997;27(5):443–94.
58. Savitz, D.A.e.a., Contaminated water supplies at Camp Lejeune: assessing potential health effects 2009: National Academies Press.
59.•• Talbott EO et al. Risk of leukemia as a result of community exposure to gasoline vapors: a follow-up study. Environmental Research. 2011;111(4):597–602. **This study suggests a possible association between chronic low-level benzene exposure due to a leaking underground storage tank and increased risk of leukemia among residents that live nearby a gas station**.
60. Sanders PF, Hers I. Vapor intrusion in homes over gasoline-contaminated ground water in Stafford, New Jersey. Ground Water Monitoring and Remediation. 2006;26(1):63–72.
61. Caprino L, Togna GI. Potential health effects of gasoline and its constituents: a review of current literature (1990–1997) on toxicological data. Environmental Health Perspectives. 1998;106(3):115.
62. U.S. Environmental Protection Agency, Design criteria for Stage 1 vapor control systems—gasoline service stations. 1975.
63. U.S. Environmental Protection Agency, Technical guidance—stage II vapor recovery systems for control of vehicle refueling emissions at gasoline dispensing facilities, Volume I: Chapters, 1991.
64. McPhee, J., Gasoline dispensing facility (GDF) balance hose permeation study, 2008, California Air Resources Board (CARB)
65. U.S. Environmental Protection Agency. Commonly asked questions about ORVR. Available from: http://www.epa.gov/otaq/regs/ld-hwy/onboard/orvrq-a.txt.
66. Musser, G. and H. Shannon, Onboard control of refueling emissions. 1986: p. SAE Technical Paper 861560.
67. Meszler Engineering Services, Stage II emission reduction benefits. Report to the Maryland Department of the Environment. 2012.
68. Federal Register, Air quality: widespread use for onboard refueling vapor recovery and stage II waiver. Final Rule by US Environmental Protection Agency. Federal Register 2012. 77(95).
69.•• Currie J et al. Environmental health risks and housing values: evidence from 1,600 toxic plant openings and closings. American Economic Review. 2015;105(2):678–709. **This study provides an empirical framework for understanding the effects and the health cost of toxic atmospheric emissions. The research design could be applied to pollution prevention at gas stations. Such analysis could provide important policy recommendation in order to mitigate this type of environmental risk**.
70. Ni J. Environmental cost and economic benefit of commercial real estate development. Working Paper: Johns Hopkins University; 2015.

71. Fong M et al. California dry cleaning industry technical assessment report. State of California Air Resources Board: Technical report; 2006.
72. Irigaray P et al. Lifestyle-related factors and environmental agents causing cancer: an overview. Biomedicine & Pharmacotherapy. 2007;61(10):640–58.
73. McGarity TO. MTBE: a precautionary tale. Harvard Environmental Law Review. 2004;28(2):281–342.
74. Powers SE et al. The transport and fate of ethanol and BTEX in groundwater contaminated by gasohol. Critical Reviews in Environmental Science and Technology. 2001;31(1):79–123.
75. Jakobsson R et al. Acute myeloid-leukemia among petrol station attendants. Archives of Environmental Health. 1993;48(4):255–9.
76. U.S. Environmental Protection Agency, EPA requires phase-out of lead in all grades of gasoline. EPA press release – November 28, 1973. 1973.
77. U.S. Energy Information Administration (EIA), MTBE, oxygenates, and motor gasoline. 2000.
78. U.S. Environmental Protection Agency. Assessment of potential health risks of gasoline oxygenated with Methyl Tertiary Butyl Ether (MTBE). Washington, DC:Office of Research and Development, U.S. EPA. 1993. EPA/600/R-93/206.

2019 Study
Vent pipe emissions from storage tanks at gas stations: Implications for setback distances

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Vent pipe emissions from storage tanks at gas stations: Implications for setback distances

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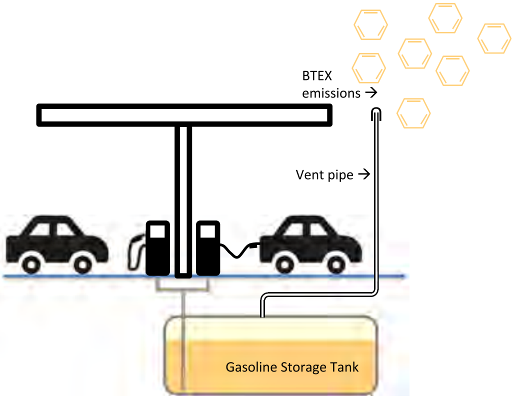
HIGHLIGHTS

- At gas stations, fuel vapors are released from storage tanks through vent pipes.
- We measured vent pipe flow rates and tank pressure at high temporal resolution.
- Vent emission factors were >10 times higher than previous estimates.
- Modeling was used to examine exceedance of benzene short-term exposure limits.

GRAPHICAL ABSTRACT



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ABSTRACT

At gas stations, fuel vapors are released into the atmosphere from storage tanks through vent pipes. Little is known about when releases occur, their magnitude, and their potential health consequences. Our goals were to quantify vent pipe releases and examine exceedance of short-term exposure limits to benzene around gas stations. At two US gas stations, we measured volumetric vent pipe flow rates and pressure in the storage tank headspace at high temporal resolution for approximately three weeks. Based on the measured vent emission and meteorological data, we performed air dispersion modeling to obtain hourly atmospheric benzene levels. For the two gas stations, average vent emission factors were 0.17 and 0.21 kg of gasoline per 1000 L dispensed. Modeling suggests that at one gas station, a 1-hour Reference Exposure Level (REL) for benzene for the general population (8 ppb) was exceeded only closer than 50 m from the station's center. At the other gas station, the REL was exceeded on two different days and up to 160 m from the center, likely due to non-compliant bulk fuel deliveries. A minimum risk level for intermediate duration (>14–364 days) benzene exposure (6 ppb) was exceeded at the elevation of the vent pipe opening up to 7 and 8 m from the two gas stations. Recorded vent emission factors were >10 times higher than estimates used to derive setback distances for gas stations. Setback distances should be revisited to address temporal variability and pollution controls in vent emissions.

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1. Introduction

In the US, approximately 143 billion gal (541 billion L) of gasoline were dispensed in 2016 at gas stations (EIA, 2017) resulting in release of unburned fuel to the environment in the form of vapor or liquid (Hilpert et al., 2015). This is a public health concern, as unburned fuel chemicals such as benzene, toluene, ethyl-benzene, and xylenes (BTEX) are harmful to humans (ATSDR, 2004). Benzene is of special concern because it is causally associated with different types of cancer (IARC, 2012). Truck drivers delivering gasoline and workers dispensing fuel have among the highest exposures to fuel releases (IARC, 2012). However, people living near or working in retail at gas stations, and children in schools and on playgrounds can also be exposed, with distance to the gas stations significantly affecting exposure levels (Terres et al., 2010; Jo & Oh, 2001; Jo & Moon, 1999; Hajizadeh et al., 2018). A meta-analysis (Infante, 2017) of three case-control studies (Steffen et al., 2004; Brosselin et al., 2009; Harrison et al., 1999) suggests that childhood leukemia is associated with residential proximity to gas stations.

Sources of unburned fuel releases at gas stations include leaks from storage tanks, accidental spills from the nozzles of gas dispensers (Hilpert & Breysse, 2014; Adria-Mora & Hilpert, 2017; Morgester et al., 1992), fugitive vapor emissions through leaky pipes and fittings, vehicle tank vapor releases when refueling, and leaky hoses, all of which can contribute to subsurface and air pollution (Hilpert et al., 2015). Routine fuel releases also occur through vent pipes of fuel storage tanks but are less noticeable because the pipes are typically tall, e.g., 4 m. These vent pipes are put in place to equilibrate pressures in the tanks and can be located as close as a few meters from residential buildings in dense urban settings (Fig. 1).

Unburned fuel can be released from storage tanks into the environment through “working” and “breathing” losses (Yerushalmi & Rastan, 2014). A working loss occurs when liquid is pumped into or out of a tank. For a storage tank, this can happen when it is refilled from a tanker truck or when fuel is dispensed to refuel vehicles (Statistics Canada, 2009) if the pressure in the storage tank exceeds the relief pressure of the pressure/vacuum (P/V) valve (EPA, 2008). P/V valve threshold pressures are typically set to around +3 and −8 in. of water column (iwc) (7.5 and −20 hPa). However, P/V valves are not always used, particularly in cold climates, as valves may fail under cold weather conditions (Statistics Canada, 2009).

Breathing losses occur when no liquid is pumped into or out of a tank because of vapor expansion and contraction due to temperature and barometric pressure changes or because pressure in the storage tank may increase when fuel in the tank evaporates (Yerushalmi & Rastan, 2014; EPA, 2008). Although delayed or redirected by the P/V valve, breathing emissions can be significant and represent an environmental and health concern (Yerushalmi & Rastan, 2014).

Fig. 1. The three vent pipes (enclosed by the red ellipse) on the right side of the convenience store of a gas station are <10 m away from the residential building. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

Stage I vapor recovery systems, put in place to prevent working losses while delivering fuel to a station, collect the vapors displaced while loading a storage tank, redirecting them into the delivery truck. Stage II vapor recovery systems minimize working losses while delivering gas from the storage tank to the customer's car. During Stage II vapor recovery, gasoline vapors can be released through the vent pipe, if the sum of the flow rates of the returned volume and of the fuel evaporating within the storage tank is greater than the volume of liquid gasoline dispensed (Statistics Canada, 2009). We refer to this scenario as pressure while dispensing (PWD). In theory, a properly designed Stage II vapor recovery system should not have working losses, although in practice this is not typically the case (McEntire, 2000).

Regulations on setback distances for gas stations are based on lifetime cancer risk estimates. Several studies have assessed benzene cancer risk near gas stations (Atabi & Mirzahosseini, 2013; Correa et al., 2012; Cruz et al., 2007; Edokpolo et al., 2015; Edokpolo et al., 2014; Karakitsios et al., 2007). Based on cancer risk estimations, the California Air Resources Board (CARB) recommended that schools, day cares, and other sensitive land uses should not be located within 300 ft. (91 m) of a large gas station (defined as a facility with an annual sales volume of 3.6 million gal = 13.6 million L or greater) (CalEPA/CARB, 2005). This CARB recommendation has not been adopted by all US states, and within states setback distances can depend on local government. Notably, CARB regulations do not account for short term exposure limits and health effects. An important limitation of existing regulations is the use of average gasoline emission rates estimated in the 90s that do not consider excursions (CAPCOA, 1997).

The main objective of this study is to evaluate fuel vapor releases through vent pipes of storage tanks at gas stations based on vent emission measurements conducted at two gas stations in the US in 2009 and 2015, including the characterization of excursions at a high temporal resolution (~minutes) and meteorological conditions at an hourly temporal resolution. In addition, we performed hourly simulations of atmospheric transport of emitted fuel vapors to inform regulations on setback distances between gas stations and adjacent sensitive land uses by comparing modeled benzene concentrations to four 60-min benzene exposure limits: an acute Reference Exposure Level (REL) for infrequent (once per month or less) exposure (WHO, 2010) and Emergency Response Planning Guidelines ERPG-1, ERPG-2 and ERPG-3 (AIHA, 2016). Finally we compared simulated benzene levels to a Minimal Risk Level (MRL) for benzene for intermediate exposure duration (14 to 364 days) (ATSDR, 2018) because that duration window includes our duration of data collection. See Table 1 for the various benzene exposure limits and issuing agencies.

2. Methods

Although we provide SI unit conversions, we report some measures in English engineering units (ft, gal, and lb) as regulatory agencies such as CARB use these units.

2.1. Sites

Data for this study were obtained from vent release measurements conducted at two gas stations as part of technical assistance to the gas stations to quantify fuel vapor losses through the vent pipes of their storage tanks. A motivation for conducting the measurements was to perform a cost-benefit analysis to compare the economic losses due to the lost fuel versus the cost of technologies that reduce the emissions. The exact location of the two gas stations is not revealed for confidentiality reasons. The gas station managers and staff who authorized the

Table 1
Benzene exposure limits, to which we compared simulation results. For unit conversion, we assumed a temperature of 25 °C, i.e., 1 ppm = 3194 μg/m^3 (CAPCOA, 1997).

Agency	Name	Value (ppb)	Value (μg/m^3)	Exposure duration
California Office of Environmental Health Hazard Assessment (OEHHA)	REL	8	26	1 h
American Industrial Hygiene Association (AIHA)	ERPG-1	50	159,700	1 h
AIHA	ERPG-2	150	479,100	1 h
AIHA	ERPG-3	1000	3,194,000	1 h
Agency for Toxic Substances and Disease Registry (ATSDR)	MRL	6	19	14 to 364 days

ERPG = Emergency Response Planning Guidelines. The primary focus of ERPGs is to provide guidelines for short-term exposures to airborne concentrations of acutely toxic, high-priority chemicals.

collection and analysis of these data have not been involved in the current manuscript.

The first gas station, "GS-MW," was located in the US Midwest and is a 24-hour operation. The study was conducted from December 2014 to January 2015 for 20 full days, and fuel sales $\dot{V}_{sales}$ were about 450,000 gal (1.7 million L) per month. Fuel deliveries to the gas station usually took place during the nighttime. The second gas station, "GS-NW," was located on the US Northwest coast and closed at night. Hours of operation were between 6:00 am and 9:30 pm on weekdays and between 7 am and 7 pm on weekends. That study was conducted in October 2009 for 18 full days, and fuel sales were $\dot{V}_{sales}$ ~700,000 gal (2.6 million L) per month.

Both gas stations are considered to be high-volume, because they dispense >3.6 million gal of gasoline (both regular and premium) per year (CalEPA/CARB, 2005), and fuel was stored in underground storage tanks (USTs), which is typical in the US. Both gas stations had Stage II vapor recovery installed using the vacuum-assist method. In that method, gasoline vapors, which would be ejected into the atmosphere as a working loss during refueling of customer vehicle tanks, are collected at the vehicle/nozzle interface by a vacuum pump. The recovered vapors are then directed via a coaxial hose back into the combined storage tank ullage (head space) of the gas station. Stage I vapor recovery was also used at both gas stations during fuel deliveries. Both sites had a 3-inch diameter (7.5 cm) single above-grade vent pipe with below-grade manifold that connected the vent lines from several USTs; the cracking pressures of the P/V valves were set to +3 and −8 iwc (+7.5 and −20 hPa).

2.2. Vent emission measurements

To quantify evaporative fuel releases through the vent pipe of a storage tank, the volumetric flow of the mixture of gasoline vapor and air was measured in the vent pipe. A dry gas diaphragm flow meter (American Meter Company, Model AC-250) was used. For each cubic foot (28 L) of gas flowing through the meter, a digital pulse was generated. Every minute, the number of pulses was read out and stored together with date and time on a data logger. Gas flow meters were obtained from a distributor calibrated and equipped with temperature compensation and a pulse meter.

To determine the time-dependent volumetric flow rate $Q(t)$ of the gasoline vapor/air mixture through the vent pipe, the time series of measured flow volumes were integrated over an averaging period (15 or 60 min) and divided by the duration of that period. I.e., $Q(t)$ is given by the number of pulses registered by the gas flow meter in a time window multiplied by 1 cubic foot and divided by the averaging time. The 15-minute averaging time was chosen to visualize time-dependent data, while the 60-minute averaging time was chosen because air pollution simulations were performed at that resolution.

Gas pressure p in the ullage of the storage tank was measured to assess vent emission patterns. For instance, releases can occur when the pressure exceeds the cracking pressure of the P/V valve in the vent pipe (the dry gas flow meter was fitted with a P/V valve on the outlet). Pressure was measured with a differential pressure sensor (Cerabar PMC 41, Endress + Hauser) every 4 s, and 2-minute average values were stored. The sensor range was scaled from −15 to +15 iwc (−37 to +37 hPa), with a full scale accuracy of 0.20%. We also obtained 15- and 60-minute averaged tank pressure data $p(t)$ where averages represent the means of the 2-minute average pressure measurements taken during each time window.

2.3. Descriptive analysis

For the 60-minute flow rate, we calculated medians and inter quartile ranges (IQRs). To illustrate diurnal fluctuations in vapor emissions, we created box plots for the 60-minute flow rate distribution that occurred during each hour of the day. Spearman correlation coefficients between the time series for pressure and flow rate were calculated to evaluate whether pressure can be used to infer vent emissions.

To estimate the mass flow rate of gasoline $\dot{m}_{gas}$ that is released through the vent pipe in the form of a mixture of gasoline vapors and fresh air, we assumed, following the protocol of a study by the California Air Pollution Control Officers Association (CAPCOA) that assessed risks from fuel emissions from gas station (Appendix D-2 (CAPCOA, 1997)), that the density of gasoline vapors in this mixture is given by $\rho_{gas}^{(v)} = 0.3 \times 65\ \text{lb} / 379\ \text{ft}^3 = 0.824\ \text{kg/m}^3$, i.e., the molar percentages of gasoline and air were 30% and 70%, respectively. Then the volumetric flow rate Q can be converted into a mass flow rate of the vaporized gasoline:

$$\dot{m}_{gas} = \rho_{gas}^{(v)}\ Q \tag{1}$$

To arrive at vent emission factors, we first calculated the mean volumetric flow rate $\overline{Q}$, and then the mean mass flow rate $\overline{\dot{m}}_{gas} = \rho_{gas}^{(v)}\ \overline{Q}$. From the latter, one can calculate the vent emission factor

$$\text{EF}_{vent} = \overline{\dot{m}}_{gas} / \dot{V}_{sales} \tag{2}$$

For EF$_{vent}$, CARB uses units of pounds of emitted gasoline vapors (also called total organic gases (TOG)) per 1000 gal dispensed, or more briefly lb/kgal where kgal stands for kilogallons.

As we were not able to measure benzene levels in the tank ullage, we assumed like the CAPCOA study (Section C) that the density of the mixture of gasoline vapors and fresh air was $\rho_{mix}^{(v)} = 1.05\ \text{lb/ft}^3 = 1.682\ \text{kg/m}^3$ and that the emitted gasoline vapor/air mixture contained 0.3% of benzene by weight (CAPCOA, 1997). Therefore, the mass flow rate of benzene through the vent pipe was estimated as follows:

$$\dot{m}_{benz} = 0.003\ \rho_{mix}^{(v)}\ Q \tag{3}$$

2.4. Air pollution modeling

We used the AERMOD Modeling System developed by the US Environmental Protection Agency (EPA) to model the dispersion of benzene vapors released into the environment through vent pipes of fuel storage tanks and from other sources (Cimorelli et al., 2005). AERMOD simulates atmospheric pollutant transport at a 1-hour temporal resolution. 3D polar grids were created with the gas station in the origin and potential receptors at different radial distances (up to 170 m) and angles (10°

increments). The grids were placed at the ground level ($z = 0$ m), in the breathing zone ($z = 2$ m), and at the 2nd floor level ($z = 4$ m) where the vent pipe emissions were assumed to occur. The topography was simplified for modeling purposes consistent with the CAPCOA study (CAPCOA, 1997), i.e., the terrain was assumed to be flat with no buildings present. Vent pipe emissions were modeled as a capped point source. Chemical reactions of benzene were not modeled, as residence times of atmospheric benzene are on the order of hours or even days (ATSDR, 2007), i.e. much longer than the travel time of benzene vapors across the 340-m diameter model domain.

For the period of time when vent emission measurements were made, we obtained meteorological data at a 1-hour temporal resolution that are representative for the geographic locations of the two gas stations. Table SI-1 provides descriptive statistics of that data. The time series were used in AERMOD to model the transport of benzene in the temporally varying turbulent atmosphere. We also used the 1-hour average time series of benzene emission rates (Eq. (3)) as an input into AERMOD.

To evaluate at each grid point whether OEHHA's acute REL or AIHA's ERPG levels were exceeded at least once, we determined maximum 1-hour average benzene concentrations that were simulated for about three weeks. To evaluate how often the OEHHA REL was exceeded at each grid point in the breathing zone, we created plots indicating the number of exceedances and the day when the maximum benzene level was observed.

To facilitate comparison to published benzene measurements around gas stations, we determined for each simulated radial distance from a gas station the mean of the average concentrations simulated for each ten degree increment on the radius around the gas station.

3. Results: vent releases

3.1. Times series of tank pressure and flow rate

Fig. 2 shows the time-series data for the volumetric flow rate Q of the gasoline vapor/air mixture through the vent pipe and tank pressure p that we collected at the two gas stations. At GS-MW, little vapor was typically released in the late night and in the very early morning, while releases were generally much higher during the daytime and evenings, presumably when more fuel was dispensed (Fig. 2a). Occasionally, no vapor releases occurred for several hours. While we do not have access to time of fuel delivery records, field visits indicate that time periods with no releases coincide with fuel deliveries. For instance, fuel delivery likely occurred on January 6 at 7 pm (see Fig. 3a; an amplification of data shown in Fig. 2a). As a result, the UST pressure dropped by about 10 hPa, far below the cracking pressure of the P/V valve. The decreased gas pressure in the ullage increased until the cracking pressure of the P/V valve was reached. A very small vapor release (~2 L/min) was observed briefly on the next day at 2 am. The vapor flow rate becomes relatively large again, ~12 L/min, only after 6 am, i.e., 11 h after fuel delivery.

Fig. 3b amplifies a major vapor release at GS-MW. The UST pressure significantly exceeded the cracking pressure of the P/V valve and rose rapidly up to 37 hPa, which coincides with vapors being released at a high flow rate (15-min average) of about 470 L/min.

At GS-NW, vapor releases followed a quite different pattern (Fig. 2b). Contrary to GS-MW, vapor releases occurred in a cyclical pattern, and tended to be higher in the late night and in the very early morning when the gas station was closed.

3.2. Statistics of vapor emissions

The average volumetric flow rate $\overline{Q}$ through the vent pipe for the entire period of time during which measurements were taken was $\overline{Q} = 7.9$ L/min for GS-MW and $\overline{Q} = 15.4$ L/min for GS-NW, which is consistent with the higher sales volume $\dot{V}_{sales}$ of GS-NW. These emissions consist of a mixture of gasoline vapors and air. Using Eq. (1), the volumetric flow rates were converted into average mass flow rates of gasoline: $\overline{m}_{gas} = 0.39$ kg/h for GS-MW and $\overline{m}_{gas} = 0.76$ kg/h for GS-NW. Using Eq. (2), we determined a vent emission factor $\mathrm{EF}_{vent} = 0.17$ kg per 1000 L = 1.4 lb/kgal for GS-MW and $\mathrm{EF}_{vent} = 0.21$ kg per 1000 L = 1.7 lb/kgal for GS-NW.

The medians (IQRs) for the 60-minute averaged flow rate Q (L/min) were 6.1 (1.9, 10.9) for GS-MW and 16.0 (12.7, 18.4) for GS-NW. For GS-MW, the mean is larger than the median, indicating a more skewed distribution of flow rates when compared to GS-NW. Also the first quartile is much lower than the median for GS-MW, indicating that there are periods of time during which little emissions occurred. Conversely, GS-NW was releasing emissions more consistently.

Fig. 4a shows boxplots illustrating the distribution of flow rate Q for each hour of the day at GS-MW. Less vapor was released between 10 pm and 4 am, even though the gas station was in operation, albeit at lower activity levels. The flow rate Q at GS-NW (Fig. 4b) had fewer outliers, and the highest outlier was an order of magnitude lower than the highest one at GS-MW. Emissions were highest between 1 and 3 am, when the gas station was closed.

The Spearman correlation coefficients between tank pressure p and vent flow rate Q were $r = 0.58$ for GS-MW and $r = 0.85$ for GS-NW. Thus, vent releases are moderately and strongly correlated with tank pressure, respectively. Table 2 summarizes statistical properties of vent emissions at the two gas stations.

4. Results: air pollution modeling

4.1. Emission sources and rates

Vent pipe emissions of benzene were modeled at a 1-hour temporal resolution as described in Section 2.4. However, they are not the sole source of gasoline emissions at gas stations. Accidental spills from nozzles regularly occur near the dispensers, “refueling losses” can occur when gasoline vapors are released from the vehicle tank during refueling due to the rising liquid levels in the tanks, fuel vapors are released from permeable dispensing hoses, and “fugitive” or leakage emissions occur with driving force derived from storage tank pressure. In Section A of Supporting material, we detail how these other emission sources were modeled. Table 3 summarizes estimated mean emission rates. Note that the vent pipe losses are much greater than other losses.

4.2. Predicted benzene levels

Fig. 5 shows for both gas stations and at each grid point the maximum 1-hour average benzene concentration observed during the simulated periods in time. Benzene levels depend significantly on elevation within a 50-meter radius around the centers of the gas stations. Close to the centers of the gas stations, benzene levels are higher at the 4-m elevation and at ground level due to vent pipe emissions, which represent the largest emission source (Table 3). Further than 50 m away from the center, the vertical concentration differences become less obvious due to dispersion causing vertical mixing of benzene vapors.

At GS-MW, the 1-hour acute REL of 26 $\mu g/m^3$ was exceeded 160 m away from the center of the gas station, at the location ($x = 158$ m, $y = 28$ m) both at ground level and in the breathing zone. At grid points with a distance >50 m from the center of the gas station, the REL was exceeded at most once (Fig. SI-1a). However, the exceedance at different grid points did not occur on the same day (Fig. SI-1b). Within the 20 days during the measurement campaign, exceedances occurred on the 4th and 13th of January.

At GS-NW, the furthest REL exceedance occurred at 50 m from the center of the gas station at the grid point ($x = -38$ m, $y = 32$ m) as

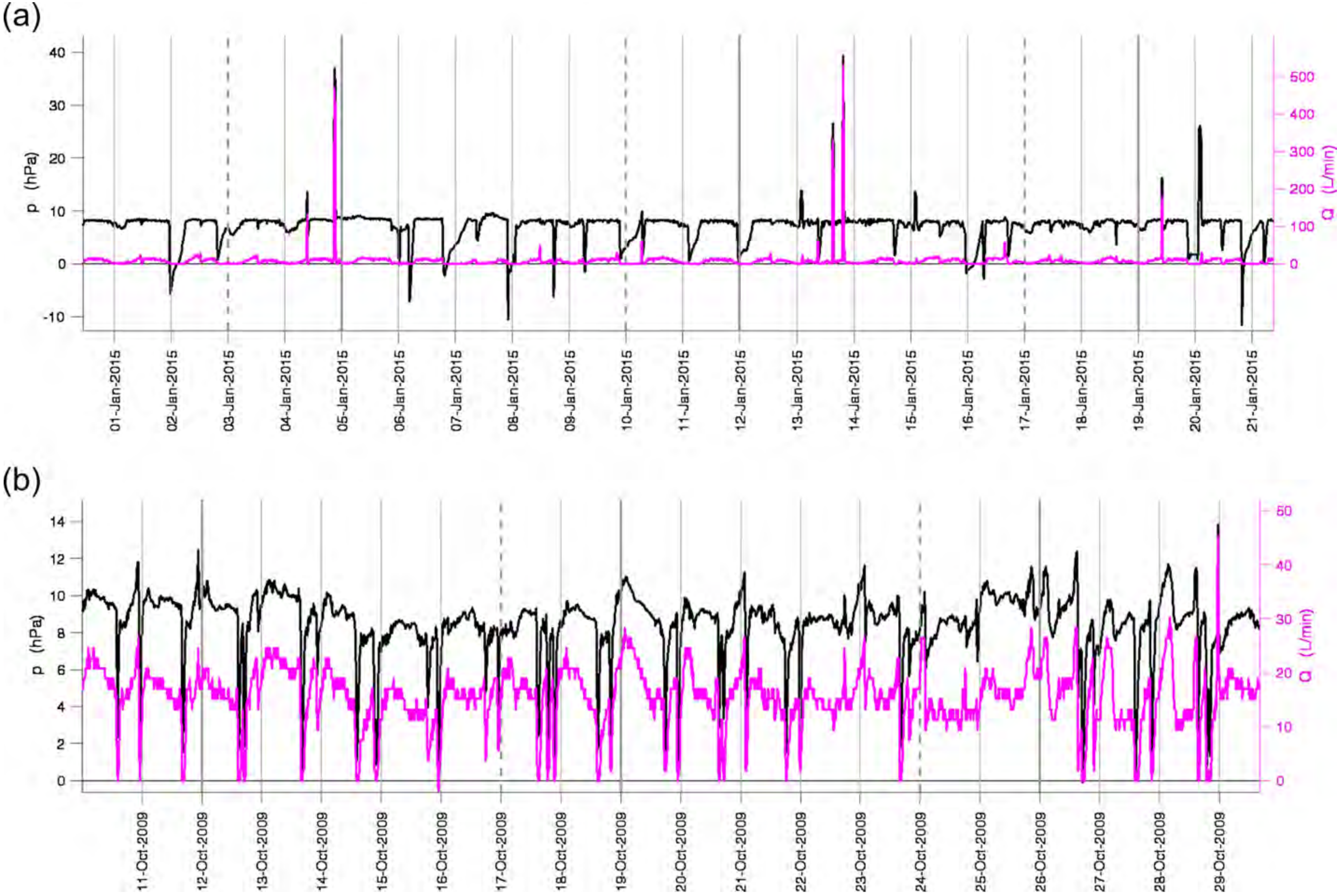

Fig. 2. Time series of ullage pressure p (left ordinate) and volumetric flow rate Q (right ordinate) for (a) GS-MW and (b) GS-NW. Horizontal tick marks indicate midnights. The vertical dashed and thick solid gray lines enclose weekends.

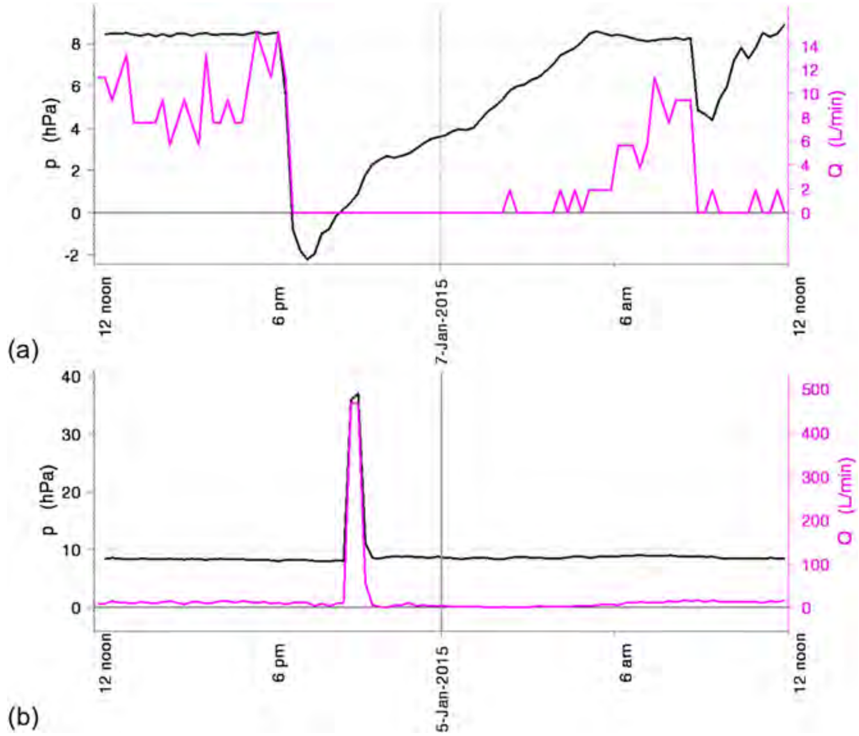

Fig. 3. Amplifications of time series data (15-minute averages) for GS-MW. (a) Tank pressure *p* became negative after fuel delivery. As a result, vent emission ceased for several hours. (b) A major vapor release (burst) likely occurred when the cracking pressure of the P/V valve was significantly exceeded at around 9 pm during a non-compliant bulk fuel delivery.

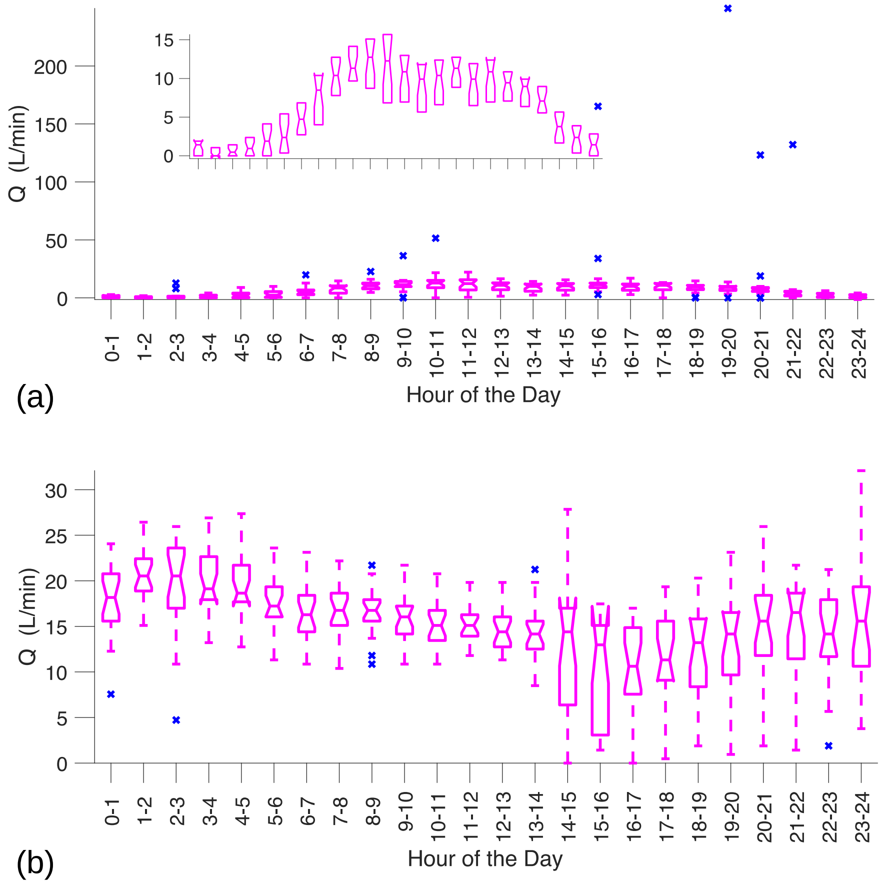

Fig. 4. Distribution of vent emissions Q observed for each hour of the day at (a) GS-MW [insert shows the IQRs of *Q*] and (b) GS-NW gas stations. In (a), outliers make it difficult to recognize variations in median hourly emissions. We therefore plotted in the inset only the IQRs. Boxes indicate median and IQR, whiskers values within 1.5 the IQR, and asterisks outliers.

Table 2
Summary of gas station characteristics and vent emissions.

	GS-MW	GS-NW	Units
Sales volume $\dot{V}_{sales}$	450,000	700,000	gal/month
Volumetric flow rates (of gasoline vapor/air mixture)			
Mean $\overline{Q}$	7.9	15.4	L/min
Median (IQR) of 60-min average	6.1 (1.9, 10.9)	16.0 (12.7, 18.4)	L/min
Maximum of 60-min average	250	32.1	L/min
Vent emission factor EF_{vent}	1.4	1.7	lb/kgal
Mass flow rates of gasoline (w/o air)			
Mean $\overline{\dot{m}}_{gas}$	0.39	0.76	kg/h
Maximum of 60-min average	12.3	1.6	kg/h
Correlation coefficient			
Between Q and p	0.58	0.85	–

shown in Fig. SI-2a. At a distance of 40 m, the REL was exceeded three times at one grid point (260° angle), and at 35 m four times at two grid points (250° and 260° angles) (Fig. SI-2b). At a distance of 20 m, the REL was exceeded at 30 (out of 36) grid points, and on nine different days.

Average benzene levels are shown in Fig. 6 for both gas stations. The MRL is exceeded at the elevation of the vent pipe opening, $z = 4$ m, up to 7 m away from for GS-MW and up to 8 m from GS-NW. Fig. 7 shows the average benzene concentration as a function of distance at an elevation of 2 m. Close to the center, benzene levels first increase and then decrease.

5. Discussion

5.1. Vent emission factors

We present unique data on vent emissions from USTs at two gas stations. Emissions can be compared to vent losses assumed by CAPCOA (CAPCOA, 1997). For a gas station with Stage I and II vapor recovery technology and a P/V valve on the vent pipe of the UST (Scenario 6B), the CAPCOA study assumed loading losses of 0.084 and breathing losses of 0.025 lb/kgal dispensed. The total loss of gasoline through the vent pipe is the sum of the two and amounts to a vent emission factor $EF_{vent}=$ 0.109 lb/kgal. Based on actual measurements in two fully functioning US gas stations, we obtained EF_{vent} values of 1.4 lb/kgal for GS-MW and 1.7 lb/kgal for GS-NW, more than one order of magnitude higher than the CAPCOA estimate. While the difference between our measurements and the CAPCOA estimates may appear surprising, it is important to consider that the CAPCOA estimates are based on relatively few measurements and some unsupported assumptions (Aerovironment, 1994), particularly with regard to uncontrolled emissions due to equipment failures or defects (Appendix A-5 (CAPCOA, 1997)).

5.2. Pressure measurements

Tank ullage pressure p was moderately to strongly positively correlated with vent flow rate Q, likely because exceedance of the cracking pressure of the P/V valve causes a vent release. Thus pressure measurements can be used to infer vent releases. Real-time detection of equipment failures and leaks via so-called in-station diagnostics systems is based on our observed correlations between p and Q.

Table 3
Mean benzene emission rates $\dot{m}_{benz}$ for the two gas stations.

Emission source	Benzene emissions (mg/s)	
Gas station	GS-MW	GS-NW
Vent pipe	0.80	1.55
Spillage	0.39	0.65
Refueling	0.41	0.69
Hose permeation	0.06	0.10
Total	1.67	2.90

5.3. Diurnal fluctuations in vent emissions

Diurnal vent emissions were quite different at the two gas stations. At GS-MW, a 24-hour operation, vent emissions were high during the daytime, presumably due to PWD. Emissions ceased at night, likely because less gasoline was dispensed and fuel deliveries with relatively cool product were frequent. Evaporative losses could also have been lower at night because the cooler delivered fuel would cause slight contraction of the liquid phase with corresponding growth in the ullage volume while at the same time lowering the vapor pressure of gasoline in the UST.

At GS-NW, vent pipe releases occurred most of the time, during the daytime when fuel was dispensed (PWD) and at night when the gas station was closed. Vent releases were higher when the gas station was closed, suggesting that during the day-time Stage II vapor recovery resulted in the injection of vapors into the storage tank that were not completely equilibrated with the liquid gasoline. During night-time, the gradual equilibration of unsaturated air in the ullage of the UST with gasoline vapors could then have caused exceedance of the cracking pressure of the P/V valve and consequently vapor release. It seems counterintuitive that less nighttime emissions occurred at the gas station where fuel was dispensed. However, while fuel is being dispensed, the outgoing liquid creates additional ullage volume, and depending on excess air ingestion rate, a negative pressure could result that lowers vent pipe emissions.

Dispensing fuel to customer vehicles and the associated Stage II vapor recovery system interact with vent emissions and can even cause vent emission during PWD, because the vacuum-assist method can negatively interfere with Onboard Refueling Vapor Recovery (ORVR) installed in customer vehicles (EPA, 2004). However, Stage II vapor recovery is not obsolete. It can be used in conjunction with ORVR to minimize exposure of gas station customers and workers to benzene due to working losses (Cruz-Nunez et al., 2003), particularly when customer vehicles are not equipped with ORVR (e.g., older vehicles, boats, motorcycles) or small volume gasoline containers are refueled. Enhanced Stage II vapor recovery technology can significantly reduce vapor emissions both at the nozzle and from UST vent pipes (CARB, 2013).

5.4. Fuel deliveries and accidental vent releases

Based on observations and interpretation of time series of the tank pressure data, it is likely that the peak vent emissions (e.g., Fig. 3b) were partly due to non-compliant bulk fuel drops where the Stage I vapor recovery system either was not correctly hooked up by the delivery driver or to hardware problems with piping and/or valves. This

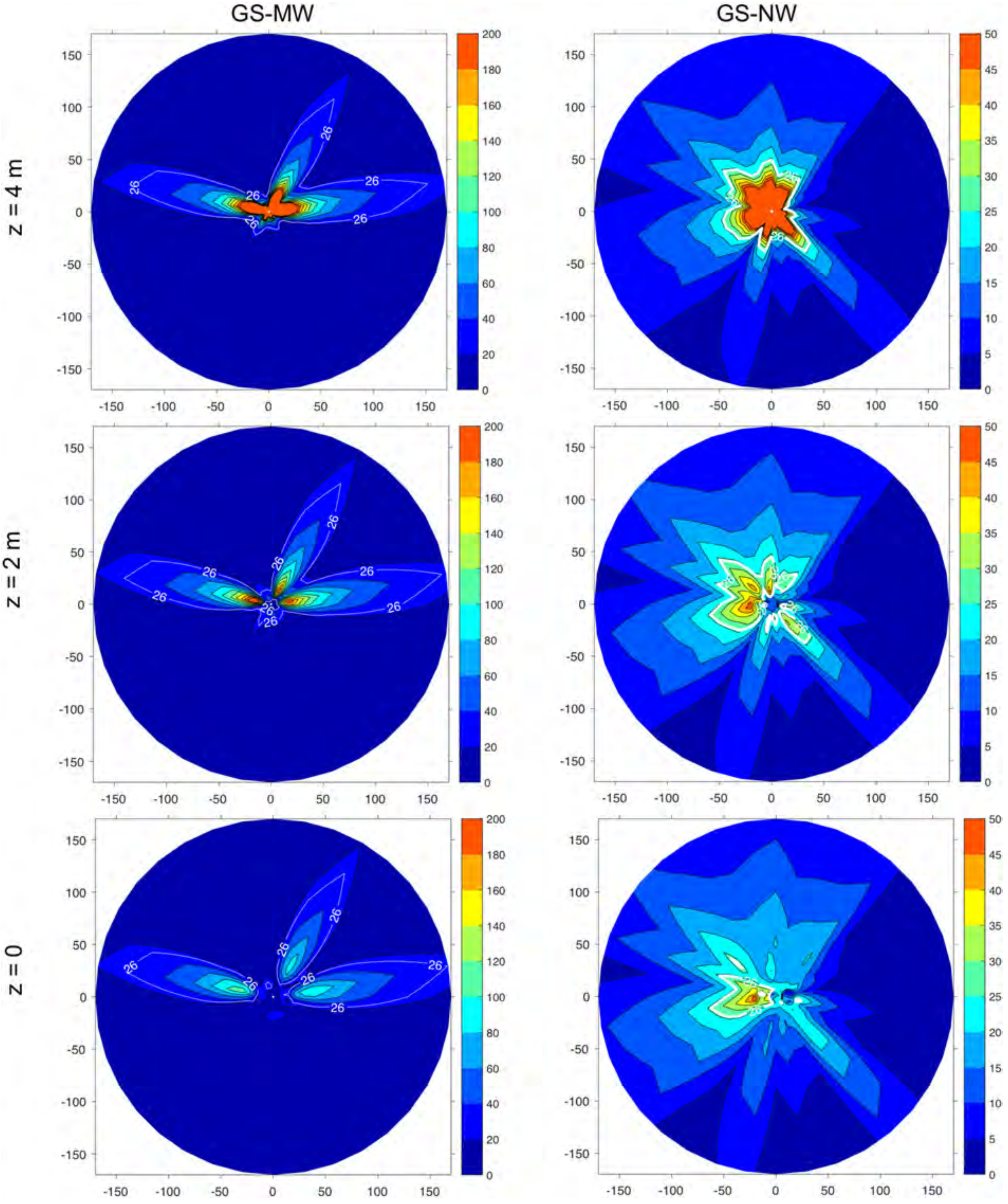

Fig. 5. Modeled maximum benzene concentrations for GS-MW and GS-NW at three different elevations *z*. The *x*- and *y*-axes indicate horizontal coordinates in meters. The color indicates benzene levels in units of μg/m^3. Left column: time series of benzene emission rates were used. Right column: average benzene emission rate was used in the modeling. The white isoline indicates OEHHA's acute REL of 26 μg/m^3 = 8 ppb.

conjecture is consistent with typical US storage tank volumes (~10,000 to 30,000 gal). Assuming that Phase I vapor recovery did not work at all and that 10,000 gal (~38,000 L) of fuel were delivered, the working loss (volume of gasoline vapor/air mixture released to the atmosphere through the vent pipe) is 38,000 L. It is also reasonable to assume that delivery lasted less than 1 h. According to Table 2, the maximum hourly flow rate through the vent pipe was 250 L/min at GS-MW, which would result in a maximum cumulative vapor release of 15,000 L within this hour. The measured maximum cumulative release underestimates the assumed working loss of 38,000 L. This could be due to a fuel delivery, which involved dropping fuel from multiple compartments of a tanker truck, with the vapor return hose not being correctly hooked up for only some of the emptied compartments.

At GS-MW, UST pressure decreased after fuel delivery (causing vent emissions to cease for several hours) during the climatic conditions prevalent during the observation period, behavior not observed at GS-NW. In practice, it is possible to observe both positive and negative pressure excursions, even during the same fuel delivery (when multiple fuel

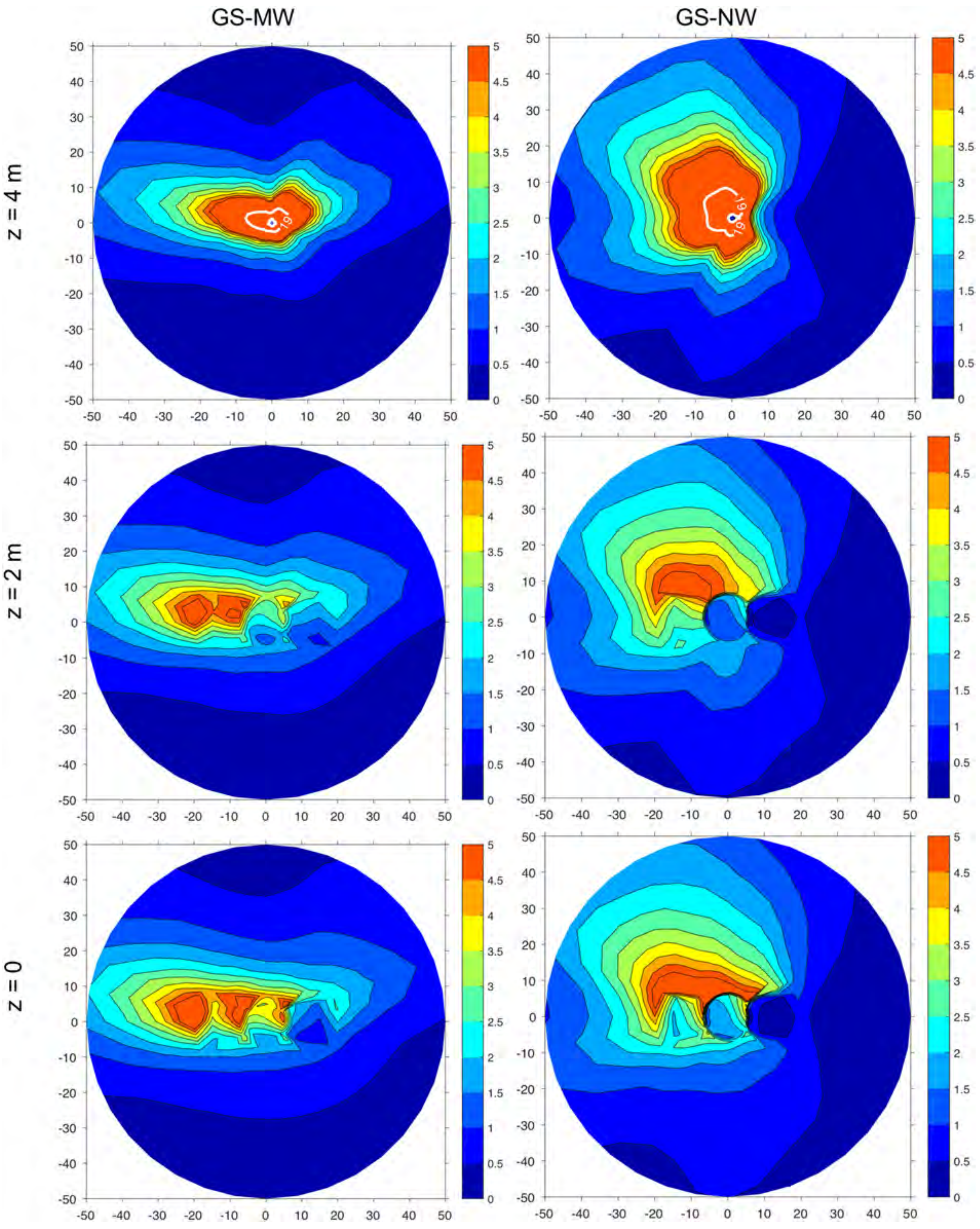

Fig. 6. Modeled average benzene concentrations for GS-MW and GS-NW at three different elevations *z*. The *x*- and *y*-axes indicate horizontal coordinates in meters. The color indicates benzene levels in μg/m^3 and the white isoline the MRL of 19 μg/m^3 = 6 ppb.

compartments of tanker trucks are unloaded), when Stage I vapor recovery is in place (personal observation by TT).

5.5. Exceedance of 1-hour exposure limits

AERMOD air pollution modeling suggests that at GS-MW the 1-hour acute REL was exceeded at one grid point 160 m (525 ft) from the center of the gas station once in 20 days (Fig. 5). This distance is larger than the 300-ft (91 m) setback distance recommended by CARB for a large gasoline dispensing facility (CalEPA/CARB, 2005). Assuming the gas station's fence line is <225 ft. (69 m) from its center (where the vent pipe was assumed to be located), our study shows that sensitive land uses at a distance further than 300 ft from the fence line of the gas station would represent a health concern despite compliance with the CARB guidelines because of non-compliance with the acute REL.

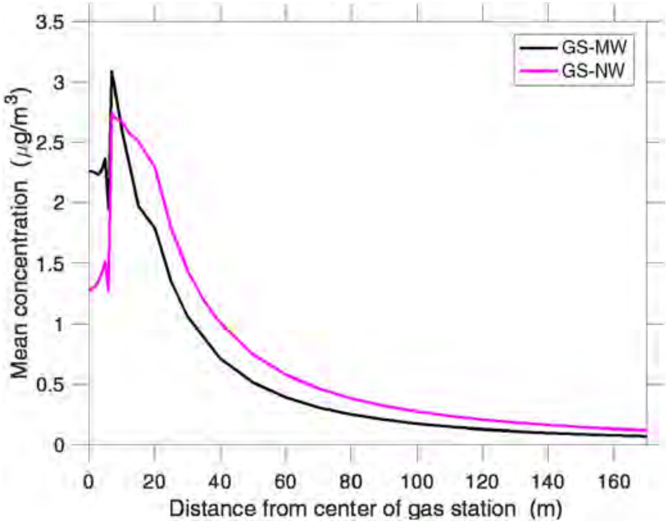

Fig. 7. Mean benzene concentrations as a function of distance from the center of the gas stations.

At any location further than 50 m from the gas station's center, the REL was exceeded at most once during the 20-day measurement campaign (Fig. SI-1a). However, exceedance occurred at several locations, and on two different days (Fig. SI-1b). E.g., at a distance of 120 m from the center, the REL was exceeded at three grid points, and the number of grid points increased with closer proximity to the gas station. This suggests that it was not just a single worst-case scenario or a single accidental vapor release that led to REL exceedance; rather exceedance may occur more frequently than is anticipated. Prevalent wind directions during the measurement campaign explained the directional patterns of exceedances (see the wind rose in Fig. SI-3a).

At GS-NW, despite its higher sales volume, the REL was exceeded only closer than 50 m from the gas station's center. However, exceedance occurred much more frequently (Fig. SI-2), likely because of the higher sales volume of GS-NW. Again, the wind rose for GS-NW (Fig. SI-3b) explains spatial patterns of REL exceedance.

None of AIHA's three ERPG levels were exceeded, meaning that individuals, except perhaps sensitive members of the public, would not have experienced more than mild, transient adverse health effects.

5.6. Average benzene levels

The initial increase in average benzene levels when moving away from the gas stations' centers (Fig. 7) is likely due to the vent emissions (at 4 m) which represent the largest benzene source, and which require a certain transport distance until they reach the 2-m level through dispersion. Further away from the gas station, benzene levels are higher for GS-NW than for GS-MW likely because of the higher sales volume of GS-NW. However, close to the center, benzene levels are higher at GS-MW. This can be attributed to the higher wind speeds at GS-NW (Table SI-1), which result in greater initial dilution of emitted pollutants in the incoming airstream and also in greater subsequent pollutant dispersion.

Modeled average benzene concentrations are generally lower (~10 μg/m^3 or less) than those measured in the surroundings of gas stations, likely because our simulations do not account for traffic-related air pollution (TRAP). For instance, a study published by the Canadian petroleum industry found average benzene concentrations of 146 and 461 ppb (466 and 1473 μg/m^3) at the gas station property boundary in summer and winter, respectively (Akland, 1993), values orders of magnitudes higher than ours. A South Korean study examined outdoor and indoor benzene concentrations at numerous residences within 30 m and between 60 and 100 m of gas stations and found median outdoor benzene concentrations of 9.9 and 6.0 μg/m^3, respectively (Jo & Moon, 1999), while we simulated benzene levels on the order of 1 μg/m^3 (Fig. 7). In a study on atmospheric BTEX levels in an urban area in Iran, the three highest BTEX levels were measured near gas stations (~150 m away); the measured benzene levels (64 ± 36, 31 ± 28, 52 ± 26 μg/m^3) were again much higher than ours simulated at that distance, likely due to TRAP. Our modeled average benzene levels at a distance of about 50 m are on the same order as background benzene levels of 1.0 μg/m^3 that were measured in 2010 in the National Air Toxics Trend Sites (NATTS) network of 27 stations located in most major urban areas in the US (Strum & Scheffe, 2016). However, our modeled levels at a distance of 170 m were 0.07 at GS-MW and 0.12 at GS-NW, a non-negligible addition to urban background levels.

At both gas stations, the MRL was exceeded at the level of the vent pipe opening in the vicinity of the gas stations, up to 7 m away from the vent pipe at GS-MW and 8 m at GS-NW. Therefore there might be an appreciable risk of adverse noncancer health effects for individuals living at the 2nd-floor level relatively close to high-volume gas stations such as GS-MW and GS-NW.

5.7. Limitations

A limitation of our study is that data were collected only in fall and winter. Results cannot be easily extrapolated to other seasons, because vent pipe emissions are seasonally dependent, e.g., due to seasonally dependent gasoline formulations and meteorological conditions. However, modeled exceedance of the OEHHA acute REL in the winter season is already of concern, because that REL was developed for once per month or less exposures.

Another limitation is that we did not directly measure benzene levels in the vent pipe, and instead made assumptions about vapor composition that were also made in the CAPCOA study (CAPCOA, 1997) of gas station emissions. In practice it may be difficult to obtain permission from gas station owners to measure benzene levels directly.

In part because we did not want to reveal the locations of the gas stations, we did not use site-specific topography information in the air dispersion modeling and instead assumed flat terrain. While this simplification results in less accurate air pollution predictions for the two sites, using a "generic" gas station is perhaps more representative of other gas station sites, and is consistent with an approach used in a previous study (CAPCOA, 1997).

Finally, our study did not predict benzene levels in indoor environments. Even though indoor air pollution levels may substantially differ from outdoor levels due to indoor sources (e.g., smoking, photocopying) (El-Hashemy & Ali, 2018), our study can still inform exposure levels in indoor environments as outdoor sources may be the main contributors to indoor air pollution, e.g., in buildings situated in urban areas and close to industrial zones or streets with heavy traffic (Jones, 1999). This is relevant to workers and customers in C-stores or other fast-food/gasoline station combination facilities.

6. Conclusions

Our study is to the best of our knowledge the first one to (1) report hourly vent emission data for gasoline storage tanks in the peer-reviewed literature and (2) use these data in hourly simulations of atmospheric benzene vapor transport. This allowed us to examine potential exceedance of short-term exposure limits for benzene. Prior studies including CAPCOA's (CAPCOA, 1997) could not do so as average emission rates were used (only meteorological data was used at an hourly resolution).

Our findings support the need to revisit setback distances for gas stations, which are based on >2-decade old estimates of vent emissions (Aerovironment, 1994). Also, CARB setback distances are based on a binary decision, related to whether the gasoline sales volume $\dot{V}_{sales}$ is >3.6 million gal per year. Our data support, however, that setback distances should be a continuous function of sales volume $\dot{V}_{sales}$ and also include the type of controls installed at the facility. Setback distances should also address health outcomes other than cancer. OEHHA's acute REL for benzene could be used to inform setback distances as it accounts for non-cancer adverse health effects of benzene and its metabolites (Budroe, 2014). ATSDR's MRL could also be considered since it is a health-based limit.

We note that CARB recommended their setback distances in 2005, presumably assuming pollution prevention technology yielding a 90% reduction in benzene emissions (CalEPA/CARB, 2005). Since then, CARB further promoted use of second-generation vapor recovery technology (Enhanced Vapor Recovery, EVR) to reduce emissions further. EVR includes technology that is supposed to prevent fuel vapors in overpressurized tanks from being expelled into the atmosphere (CARB, 2017). To that end, "bladder tanks" have been proposed, into which the gasoline vapor/air mixture is directed as the pressure in the combined ullage space of the storage tank increases, and from which the mixture is redirected into the fuel storage tanks if the ullage pressure becomes negative (when fuel is dispensed). The challenge with such a system is to ensure that the bladder tank capacity is not exceeded by the fuel evaporation rate. Alternatively, fuel vapor release can be reduced by processing the fuel/air mixture through either a semi-permeable membrane which selectively exhausts clean air and returns enriched fuel vapor (Semenova, 2004) or an activated carbon filter which adsorbs hydrocarbons (and water vapor) and exhausts air into the atmosphere, or by combusting the fuel/air mixture which would otherwise be released through the P/V valve. Therefore, current CARB setback distances might be adequate for gas stations in California but less so for the other 49 US states, and other countries—depending on pollution prevention technology requirements.

The larger areal extent of modeled REL exceedance at GS-MW is due to "accidental" releases of gasoline vapors. Even though regulations appear generally not to be driven by accidental releases, at GS-NW such releases likely led on two different days to REL exceedances at distances beyond CARB's recommended setback distances. Policies should address accidental fuel vapor releases that depending on pollution prevention technology (here Stage I vapor recovery) and its proper functioning can occur on a frequent basis (twice at GS-MW within about three weeks).

In future work, potential exceedance of other shorter-term exposure limits should be examined, e.g., the 15-minute short-term exposure limits (STELs) and the 8-hour time-weighted averages (TWAs) used for occupational exposures.

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Competing financial interest declaration

TT directs a company (ARID), which develops technologies for reducing fuel emissions from gasoline-handling operations. AMR, BAM and MH have no conflicts of interests to declare.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2018.09.303.

References

Adria-Mora, B., Hilpert, M., 2017. Differences in infiltration and evaporation of diesel and gasoline droplets spilled onto concrete pavement. Sustainability 9 (7). https://doi.org/10.3390/su9071271.

Aerovironment, 1994. I. Underground Storage Tank Vent Line Emissions form Retail Gasoline Outlets. Prepared for WSPA (AV-FR-92-01-204R2).

AIHA, 2016. ERPG/WEEL Handbook. Current ERPG® Values (2016). American Industrial Hygiene Association, p. 2016.

Akland, G.G., 1993. Exposure of the general population to gasoline. Environ. Health Perspect. 101 (Suppl. 6), 27–32 (Epub 1993/12/01. PubMed PMID: 8020446; PMCID: PMC1520004).

Atabi, F., Mirzahosseini, S.A., 2013. GIS-based assessment of cancer risk due to benzene in Tehran ambient air. Int. J. Occup. Med. Environ. Health 26 (5), 770–779. https://doi.org/10.2478/s13382-013-0157-4 (Epub 2014/01/28, PubMed PMID: 24464541).

ATSDR, 2004. Interaction Profile for: Benzene, Toluene, Ethylbenzene, and Xylenes (BTEX). Agency for Toxic Substances and Disease Registry.

ATSDR, 2007. Toxicological Profile for Benzene. Agency for Toxic Substances and Disease Registry (CAS#: 71-43-2).

ATSDR, 2018. Minimal Risk Levels (MRLs): Agency for Toxic Substances and Disease Registry. Available from: https://www.atsdr.cdc.gov/mrls/index.asp (May 24, 2018).

Brosselin, P., Rudant, J., Orsi, L., Leverger, G., Baruchel, A., Bertrand, Y., et al., 2009. Acute childhood leukaemia and residence next to petrol stations and automotive repair garages: the ESCALE study (SFCE). Occup. Environ. Med. 66 (9), 598–606.

Budroe, J., 2014. Notice of adoption of revised reference exposure levels for benzene: Office of Environmental Health Hazard Assessment (California, US). Available from: https://oehha.ca.gov/air/crnr/notice-adoption-revised-reference-exposure-levels-benzene.

CalEPA/CARB, 2005. Air Quality and Land Use Handbook: A Community Health Perspective: California Environmental Protection Agency & California Air Resources Board.

CAPCOA, 1997. Gasoline Service Station Industrywide Risk Assessment Guidelines. Toxics Committee of the California Air Pollution Control Officers Association (CAPCOA).

CARB, 2013. Revised Emission Factors for Gasoline Marketing Operations at California Gasoline Dispensing Facilities. California Air Resources Board, Monitoring and Laboratory Division.

CARB, 2017. Public workshop to discuss: overpressure conditions at gasoline dispensing facilities equipped with underground storage tanks and phase ii enhanced vapor recovery including in-station diagnostic systems. December 12–13, 2017 Diamond Bar & Sacramento, CA California Air Resources Board. Available from: https://www.arb.ca.gov/vapor/op/wrkshps/dec2017op_vr_pres.pdf.

Cimorelli, A.J., Perry, S.G., Venkatram, A., Weil, J.C., Paine, R.J., Wilson, R.B., et al., 2005. AERMOD: a dispersion model for industrial source applications. Part I: general model formulation and boundary layer characterization. J. Appl. Meteorol. 44 (5), 682–693.

Correa, S.M., Arbilla, G., Marques, M.R.C., Oliveira, K.M.P.G., 2012. The impact of BTEX emissions from gas stations into the atmosphere. Atmos. Pollut. Res. 3 (2), 163–169.

Cruz, L., Alves, L., Santos, A., Esteves, M., Gomes, Í., Nunes, L., 2007. Assessment of BTEX concentrations in air ambient of gas stations using passive sampling and the health risks for workers. J. Environ. Prot. 8, 12–25.

Cruz-Nunez, X., Hernandez-Solis, J.M., Ruiz-Suarez, L.G., 2003. Evaluation of vapor recovery systems efficiency and personal exposure in service stations in Mexico City. Sci. Total Environ. 309 (1–3), 59–68. https://doi.org/10.1016/s0048-9697(03)00048-2.

Edokpolo, B., Yu, Q.J., Connell, D., 2014. Health risk assessment of ambient air concentrations of benzene, toluene and xylene (BTX) in service station environments. Int. J. Environ. Res. Public Health 11 (6), 6354–6374 (PubMed PMID: PMC4078583).

Edokpolo, B., Yu, Q.J., Connell, D., 2015. Health risk characterization for exposure to benzene in service stations and petroleum refineries environments using human adverse response data. Toxicol. Rep. 2, 917–927.

EIA, 2017. U.S. product supplied of finished motor gasoline: U.S. Energy Information Administration. Available from: http://www.eia.gov/dnav/pet/hist/LeafHandler.ashx?n=pet&s=mgfupus1&f=m.

El-Hashemy, M.A., Ali, H.M., 2018. Characterization of BTEX group of VOCs and inhalation risks in indoor microenvironments at small enterprises. Sci. Total Environ. 645, 974–983.

EPA, 2004. Stage II Vapor Recovery Systems Issues Paper. U.S. EPA. Office of Air Quality Planning and Standards. Emissions Monitoring and Analysis Division. Emissions Factors and Policy Applications Group (D243-02).

EPA, 2008. Transportation and marketing of petroleum liquids. Environmental Protection Agency. Petroleum Industry vol. I (Chapter V, AP 42).

Hajizadeh, Y., Mokhtari, M., Faraji, M., Mohammadi, A., Nemati, S., Ghanbari, R., et al., 2018. Trends of BTEX in the central urban area of Iran: a preliminary study of photochemical ozone pollution and health risk assessment. Atmos. Pollut. Res. 9 (2), 220–229.

Harrison, R.M., Leung, P.L., Somervaille, L., Smith, R., Gilman, E., 1999. Analysis of incidence of childhood cancer in the West Midlands of the United Kingdom in relation to proximity to main roads and petrol stations. Occup. Environ. Med. 56 (11), 774–780.

Hilpert, M., Breysse, P.N., 2014. Infiltration and evaporation of small hydrocarbon spills at gas stations. J. Contam. Hydrol. 170, 39–52.

Hilpert, M., Mora, B.A., Ni, J., Rule, A.M., Nachman, K.E., 2015. Hydrocarbon release during fuel storage and transfer at gas stations: environmental and health effects. Curr. Environ. Health Rep. 2 (4), 412–422. https://doi.org/10.1007/s40572-015-0074-8.

IARC, 2012. IARC Monographs on the Evaluation of Carcinogenic Risks to Humans. vol. 100F Available from:. http://monographs.iarc.fr/ENG/Monographs/vol100F/ (December 24, 2017).

Infante, P.F., 2017. Residential proximity to gasoline stations and risk of childhood leukemia. Am. J. Epidemiol. 185 (1), 1–4.

Jo, W.K., Moon, K.C., 1999. Housewives' exposure to volatile organic compounds relative to proximity to roadside service stations. Atmos. Environ. 33 (18), 2921–2928. https://doi.org/10.1016/s1352-2310(99)00097-7.

Jo, W.K., Oh, J.W., 2001. Exposure to methyl tertiary butyl ether and benzene in close proximity to service stations. J. Air Waste Manage. Assoc. 51 (8), 1122–1128. https://doi.org/10.1080/10473289.2001.10464339.

Jones, A.P., 1999. Indoor air quality and health. Atmos. Environ. 33 (28), 4535–4564.

Karakitsios, S.P., Delis, V.K., Kassomenos, P.A., Pilidis, G.A., 2007. Contribution to ambient benzene concentrations in the vicinity of petrol stations: estimation of the associated health risk. Atmos. Environ. 41 (9), 1889–1902.

McEntire, B.R., 2000. Performance of Balance Vapor Recovery Systems at Gasoline Dispensing Facilities. San Diego Air Pollution Control District.

Morgester, J.J., Fricker, R.L., Jordan, G.H., 1992. Comparison of spill frequencies and amounts at vapor recovery and conventional service stations in California. J. Air Waste Manage. Assoc. 42 (3), 284–289.

Semenova, S.I., 2004. Polymer membranes for hydrocarbon separation and removal. J. Membr. Sci. 231 (1–2), 189–207.

Statistics Canada, 2009. Gasoline evaporative losses from retail gasoline outlets across Canada: environment accounts and statistics analytical and technical paper series. Available from: http://www.statcan.gc.ca/pub/16-001-m/2012015/part-partie1-eng.htm.

Steffen, C., Auclerc, M.F., Auvrignon, A., Baruchel, A., Kebaili, K., Lambilliotte, A., et al., 2004. Acute childhood leukaemia and environmental exposure to potential sources of benzene and other hydrocarbons; a case-control study. Occup. Environ. Med. 61 (9), 773–778. https://doi.org/10.1136/oem.2003.010868.

Strum, M., Scheffe, R., 2016. National review of ambient air toxics observations. J. Air Waste Manage. Assoc. 66 (2), 120–133. https://doi.org/10.1080/10962247.2015.1076538 (1995, PubMed PMID: 26230369, Epub 2015/08/01).

Terres, I.M.M., Minarro, M.D., Ferradas, E.G., Caracena, A.B., Rico, J.B., 2010. Assessing the impact of petrol stations on their immediate surroundings. J. Environ. Manag. 91 (12), 2754–2762. https://doi.org/10.1016/j.jenvman.2010.08.009.

WHO, 2010. WHO Guidelines for Indoor Air Quality: Selected Pollutants. World Health Organization, Geneva.

Yerushalmi, L., Rastan, S., 2014. Evaporative losses from retail gasoline outlets and their potential impact on ambient and indoor air quality. In: Li, A., Zhu, Y., Li, Y. (Eds.), Proceedings of the 8th International Symposium on Heating, Ventilation and Air Conditioning. Indoor and Outdoor Environment Vol. 1. Springer Berlin Heidelberg, Berlin, Heidelberg, pp. 13–21.

2020 Study

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Gasoline Vapor Emissions During Vehicle Refueling Events in a Vehicle Fleet Saturated With Onboard Refueling Vapor Recovery Systems: Need for an Exposure Assessment

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Background: Gasoline contains large proportions of harmful chemicals, which can be released during vehicle refueling. Onboard Refueling Vapor Recovery (ORVR) can reduce these emissions, but there is limited research on the system's efficacy over time in an actual vehicle fleet. The aims of this study are: (1) determine the feasibility of using an infrared camera to view vapor emissions from refueling; (2) examine the magnitude of refueling-related emissions in an ORVR-saturated fleet, to determine need for an exposure-assessment.

Methods: Using an infrared camera optimized for optical gas imaging of volatile organic chemicals, refueling was recorded for 16 vehicles at six gas stations. Pumps were inspected for damage, refueling shut-off valve functioning, and presence of Stage II Vapor Recovery. Vehicle make/model and age were recorded or estimated.

Results: Vapor emissions were observed for 14 of 16 vehicles at each station, with severity varying substantially by vehicle make/model and age. Use of an infrared camera allowed for identification of vapor sources and timing of release, and for visualizing vapor trajectories.

Discussion: Notably emissions occurred not only at the beginning and end of refueling but also throughout, in contrast to a prior study which did not detect increases in atmospheric hydrocarbon levels mid-refueling. Future studies are vitally needed to determine the risk to individuals during typical refueling in an ORVR saturated vehicle fleet. We recommend comprehensive exposure-assessment including real-time monitoring of emitted volatile organic compounds paired with infrared gas-imaging and measurement of internal dose and health effects of gas station customers.

Keywords: gasoline, environmental exposure, vehicle refueling, volatile organic compounds, gas station

INTRODUCTION

Gasoline is a complex mixture of many chemicals, several of which are known to adversely affect human health. Of particular concern are volatile aromatic hydrocarbons, including benzene, toluene, ethylbenzene, and xylene (BTEX group), which may be released during vehicle refueling (1, 2). For example, benzene is a known human carcinogen and is associated with multiple health problems, including respiratory, nervous system, and immunological conditions (3). In addition, studies evaluating non-cancer outcomes have found decreased red blood cell counts, hemoglobin, and hematocrit levels in gas station attendants (4). While some studies have evaluated exposures to gasoline from vehicle refueling specifically (5–7), to our knowledge, few have been completed in the past decade. It is essential that such studies are repeated frequently and in varied geographic locations, as fuel composition, weather, climate, and pollution control strategies all impact individual exposures and can change over time.

In the United States (US), changes in regulations outlining gasoline vapor recovery during vehicle refueling have made this an especially pressing question. During refueling, gasoline vapor in a vehicle's tank is pushed into the atmosphere by the rising liquid gasoline level in the tank—unless a vapor recovery system is in place. From 1998 to 2006, the US Environmental Protection Agency (EPA) rolled out a requirement that nearly all newly manufactured vehicles be equipped with onboard refueling vapor recovery (ORVR) systems (8), which function by directing vaporized gasoline into a canister on the vehicle, thereby substantially reducing escape of vapors into the atmosphere. Briefly, this requirement was rolled out in stages, first for light duty vehicles (1998: 40% of new vehicles, 1999: 80%, 2000: 100%), then for light duty trucks and vans (2001: 40%, 2002: 80%, 2003: 100%), and finally for heavier light duty trucks (2004: 40%, 2005: 80%, 2006: 100%) and trucks with a >10,000 pounds gross vehicle weight rating (100% by 2006). By 2006, nearly all new gas-powered vehicles with <14,000 pound gross vehicle weight rating were required to have ORVR systems (8). In contrast, Stage II vapor recovery systems, which are used on gasoline pumps themselves, direct vaporized gasoline into gas station underground storage tanks through systems on the pumps. In 2012, the EPA determined that the US vehicle fleet was sufficiently saturated with ORVR that states could allow the removal of Stage II systems (8), thus making vapor recovery during refueling primarily dependent on ORVR systems.

Despite this change in regulations, limited information on the efficiency of ORVR systems is available, although the US EPA suggests they are 98% efficient and require minimal maintenance (8). A German study found no measurable increases in atmospheric hydrocarbon concentrations in a Sealed Housing for Emissions Determination (SHED) in which an ORVR-equipped vehicle was placed during refueling, although increases were detected at the beginning and end of refueling (9). Even though a study of presumably non-ORVR equipped vehicles in Mexico found older vehicles to have more evaporative emissions than newer ones (10), to the best of our knowledge, no assessment of the continuous functioning of ORVR systems to reduce emissions during vehicle refueling over the course of a vehicle's lifetime, within the conditions of an actual vehicle fleet, has been completed. It is possible that as vehicles age, hoses, seals, and other parts of the gas tank and ORVR system degrade, resulting in increased vapor emissions during refueling. Additionally, while some studies (6, 7) evaluated exposure to gasoline vapors during vehicle refueling in the US, finding evidence of benzene in blood and exhaled breath samples, those studies were completed before saturation of the US vehicle fleet with ORVR systems, and are thus likely over-estimates of exposures that may occur with ORVR systems. It is not currently known whether the amount of vapors today's population is exposed to would have similar, if any, effects.

Past studies assessing exposure from vehicle refueling used aluminum tubes as passive samplers (7) and sorbent tubes attached to pumps (6) to quantify exposure to gasoline vapors, positioned in the breathing zone of participants. However, such methods may not be able to detect the lower levels of exposure anticipated from a vehicle fleet with a 98% efficient ORVR system. Additionally, while these methods quantify environmental exposure to vapors during refueling, they are not easily used for source identification or to capture the dispersion and movement of vapors at the station. It is also not possible to use these devices to determine when during a refueling event vapors are more likely to be released (i.e., at the end vs. throughout), information which can help determine the cause of vapor release. Use of other technologies, such as an infrared camera optimized for visualizing compounds present in petroleum products, is needed to determine the sources of vapors during refueling (i.e., from exhaust, the vehicle tank, or the pump nozzle) and how they move through space. Such cameras are also fine-tuned to detect very small amounts of vapors, and thus may be invaluable in determining if exposure to gasoline vapors is occurring from ORVR equipped vehicles, warranting a more involved exposure-assessment.

Research on the functioning of ORVR in the actual US vehicle fleet over time, and thus an understanding of the quantity of vapors individuals may still be exposed to, is limited. Additionally, the tools traditionally used to assess exposure to vapors during vehicle refueling do not give a complete picture, as they lack the ability to determine vapor sources and movement. With this pilot study, we aim to determine the plausibility and usefulness of conducting a full exposure-assessment for exposures to gasoline vapors during vehicle refueling, in a vehicle fleet dependent on ORVR for vapor recovery. The objectives of this pilot study are to (1) determine the feasibility of qualitatively capturing fuel vapor emissions from vehicle refueling events in New York City (NYC) using a FLIR infrared camera designed specifically to detect volatile organic compounds present in petroleum products, and to (2) examine the magnitude of fuel vapor emissions over a range of different vehicle/ORVR system ages as a precursor to assessing the continuous functioning of ORVR systems over the lifetime of a vehicle in the actual US vehicle fleet.

MATERIALS AND METHODS

Study Overview

A convenience sample of gas stations in Northern Manhattan, NYC, was selected for vapor release monitoring. At each gas station, a study member approached individuals just before they began refueling their vehicles and asked for verbal permission to record their vehicle tanks as the vehicle was refueled. This study is not human subjects research, as no information about individuals was obtained, and is thus not subject to IRB oversight.

A total of six gas stations were visited over the course of a single winter day. Three vehicle refueling events were recorded at each station, with the exception of one station where an attendant was present. For this station, only one vehicle refueling event was recorded. In total, $n = 16$ refueling events were recorded.

Data Collection

An infrared camera optimized for optical gas imaging of volatile organic chemicals (FLIR model GF320; described below) and frequently used to detect leaks in petroleum refining operations, was used to record the fuel pump nozzle and external vehicle fuel tank filler pipe during each refueling session. In addition, researchers visually inspected gasoline pumps for hose damage, refueling shut-off valve functioning, and presence of Stage II Vapor Recovery systems. Researchers recorded the make and model of the vehicle when it was visible on the outside of the automobile, while year was estimated using photographs of the vehicle. Year was estimated by searching for images of the vehicle make and model, and comparing different years, especially the front and rear bumpers and headlight shape, to those shown in the photographs. When researchers could not definitively determine the year of the vehicle, the midpoint of the plausible year range was used. Vehicles were assigned a type based on the EPA Vehicle Classification system.

Overview of FLIR Infrared Camera

The FLIR model GF320 infrared camera can detect 20 gases, including: 1-pentene, benzene, butane, ethane, ethanol, ethylbenzene, ethylene, heptane, hexane, isoprene, m-xylene, methane, methanol, methyl ethyl ketone, MIBK, octane, pentane, propane, propylene, and toluene (FLIR Systems Inc., 2017). The camera is tuned to detect very small spectral ranges, so that it can selectively visualize specific compounds that absorb or emit electromagnetic energy at that spectral range. A narrow bandpass filter is used to ensure that only gases with a strong signal in the specified infrared range are detected, and other components of the camera are built to emit very little energy, to reduce the signal-to-noise ratio. The manufacturer does not provide estimates of limits of detection of their camera, but we found that the GF320 can detect quite small vapor leakage rates, e.g., gas emissions from an unignited pocket lighter in outdoor atmospheric environments imaged from a distance of at least 2 m.

Qualitative and Statistical Analysis

To determine how representative our convenience sample is of New York State and New York City vehicle fleet ORVR saturation, we used New York State's publicly available Vehicle, Snowmobile, and Boat Registrations database to calculate the proportion of registered vehicles in both the state and city that were gasoline powered and manufactured in 2006 or later (out of all gasoline powered vehicles), the year the EPA suggests that "essentially all" new gas-powered vehicles <14,000 pounds were manufactured with ORVR systems (8). We compared this to the proportion of ORVR equipped vehicles in our sample. In addition, we compared the median vehicle manufacturing age in our sample to that of registered vehicles in New York State and City.

Each infrared video was reviewed to identify the presence and magnitude of vaporized gasoline emitted during a refueling session. An overall qualitative description of each video was created, and patterns of vapor emission were identified and assigned to each session. Vapor origin (i.e., ambient vapors vs. vapors from the vehicle fuel tank) and the timing of vapor release was reviewed in all sessions. Representative video frames of "typical" emissions for each vehicle were extracted from the middle and end of each refueling session. The vapor plume was delineated using the brush feature in Microsoft Paint based on repeated observations of the videos, and not just a single frame, as it is difficult to identify the plume from a static image.

Exploratory statistical analysis was conducted in R version 3.5.1 (11). A logistic model was fit to obtain an association between estimated vehicle age and presence of vapor release during the middle of vehicle refueling, operationalized as a binary variable. Due to the small sample size no covariates were included in the model.

Figures were created with the tidyverse package in R (12), as well as with Inkscape (www.inkscape.org) and MATLAB (The MathWorks Inc., 2010).

RESULTS

A total of 16 refueling events at six gas stations were recorded. Our convenience sample was fairly representative of the estimated ORVR penetration proportion in New York State and City vehicles: according to EPA regulations 94% of our sample should have been equipped with ORVR, while for both New York State and City, we estimate that at least 81% of registered vehicles should have been equipped with ORVR. The median manufacturing year of our sample was 2013, the same as that for New York State and City.

Table 1 provides details about gas stations and vehicles. Of the six stations, only one had a Stage II vapor recovery system, and four had liquid gasoline leaking around the hose joints. Estimated vehicle age ranged from 1 to 32 years (manufacturing years 1987–2018), and several vehicle types (e.g., SUV, mid-size car) were represented in the sample. For 15 out of 16 vehicles, vehicle age and type combination indicated they were required to contain ORVR systems. The average refueling length was 86 s. Ambient temperature ranged from 33 to 41°F (0.5–5°C).

The infrared camera was able to detect gasoline vapors during vehicle refueling. In addition, evaluation of the video files allowed researchers to identify vapor sources, pinpoint the time of vapor release during each video, and to see how the vapors moved after being emitted.

TABLE 1 | Characteristics of gas stations and vehicle refueling events.

Gas station ID	Stage II vapor recovery system	Hose joints	Vehicle ID	EPA vehicle size classification	Estimated model year	ORVR mandate*	Length of refueling (s)
2	None	No leakage	29	Minicompact car	2014	Yes	66
			30	Midsize car	2005	Yes	88
			32	Standard sport utility vehicle	2013	Yes	88
3	None	Leakage	33	Midsize car	2006	Yes	76
			34	Mid-size car	2018	Yes	78
			35	Small sport utility vehicle	2013	Yes	84
4	None	Leakage	36	Mid-size car	2008	Yes	131
			37	Standard sport utility vehicle	2018	Yes	133
			38	Standard sport utility vehicle	2015	Yes	71
8	Vacuum assist	Leakage	41	Compact car	2005	Yes	72
			42	Midsize car	2016	Yes	122
			43	Midsize car	2008	Yes	66
9	None	Leakage	44	Standard sport utility vehicle	2004	Yes	56
			45	Large car	1987	No	110
			46	Midsize car	2015	Yes	106
7	None	No leakage	47	Minivan	2013	Yes	32

**Indicates whether 100% of new vehicles were required to have included ORVR systems for the specific manufacturing year and vehicle type (i.e., light duty vehicle, light duty truck, and van, heavier light duty trucks, etc.).*

Fuel vapor emissions were observed for 14 out of 16 vehicles and at every gas station. The single vehicle older than ORVR manufacturing mandates in the US clearly had much larger refueling vapor emissions than the newer vehicles. However, the majority of newer vehicles also had substantial fuel vapor emissions, particularly at the end of refueling. Qualitative descriptions of each refueling event are provided in **Table 2**. Six overall patterns of vapor emission were identified: no vapor release (one vehicle), ambient vapors only (one vehicle), release toward the end of refueling (two vehicles), release when nozzle was withdrawn (three vehicles), release toward the end of refueling and after nozzle was withdrawn (six vehicles), and near continuous vapor release (three vehicles). **Figure 1** shows the number of vehicles in each category, and the years of the vehicles' manufacture. The three vehicles with near continuous vapor release were estimated to be 5, 11, and 32 years old. Of note, all vehicles that emitted vapors at any point during the refueling session also did so at the end of the refueling session.

Representative video frames from the middle and end of each refueling session are available in the **Supplementary Material** (two frames per vehicle). In **Figure 2**, examples from each of the six vapor emission patterns are shown, with gasoline vapor plumes delineated in blue in each frame, and vehicle IDs in the top right corner. For example, for the "release when nozzle withdrawn" category, the representative screenshot during the middle of the refueling session does not show any vapors, however, at the end of the session, vapors can be seen spilling out around the pump nozzle and the vehicle fuel tank opening. The range of emission magnitude can be seen from the various sample frames. Full video recordings for each refueling event are available at the following link: https://github.com/jenni-shearston/Vehicle_Refueling_Videos.

Results from the exploratory logistic regression were not significant, as there were not enough observations to detect an association ($n = 16$; yes release [$n = 3$]/no release [$n = 13$]). The model suggested that a 1 year increase in estimated vehicle age was associated with a 1.15 increase in likelihood of emitting vapors during the middle of refueling (95% CI = 0.97, 1.51), but this result is likely driven by the results for the 32 years old vehicle, which was much older than the rest of the vehicle population.

DISCUSSION

This work highlights the value of using an infrared camera to compliment more traditional methods of exposure measurement for determining potential health risks from vehicle refueling, and visually highlights the sometimes large amounts of fuel vapor emissions that occur even within an ORVR saturated vehicle fleet.

A FLIR camera allowed us to identify the source of the vapors; for example, in one video (Vehicle ID 44) vapors can be seen, but they do not originate from the pump nozzle or the vehicle tank. Of note, we observed leaking gasoline around the hose joints at this station (Station 9). For all other videos, vapors are clearly seen coming out of the pump nozzle, vehicle tank, or both. This allows for the differentiation of sources of vapor exposure, crucial information needed to intervene on exposures at gas stations generally, or to determine how effective ORVR is at minimizing vapor outflow. In addition, use of the infrared camera allowed us to confirm that vapors were emitted in a location where an individual filling up their gas tank might breathe them in (the "breathing zone"), and to visualize the dispersion and movement of the vapors. The infrared camera also made it possible to pinpoint when during a refueling session

TABLE 2 | Qualitative description and overall patterns of vehicle refueling events.

Vehicle ID	Qualitative description	Overall pattern
29	Some gasoline vapor can be seen escaping into the atmosphere from the beginning of the refueling event, continuing throughout the duration of refueling. At around 0:00:41, a larger amount of vapor is seen escaping from the vehicle tank, generally increasing in amount until the end of the refueling session	Near continuous vapor release
30	No vapors are seen escaping into the atmosphere until more than a minute of refueling has passed (0:01:13), after which a large amount of vapor escapes as the vehicle tank presumably reaches full	Release toward end of refueling
32	Minimal vapor was released into the atmosphere throughout the duration of the refueling event. At the very end of refueling, as the pump is removed from the tank, a small amount of vapor can be seen escaping	Release toward end of refueling and after nozzle withdrawn
33	No vapors are seen escaping from the vehicle tank until the end of refueling, around 0:01:13, after which a large amount of vapor escapes, presumably as the tank reaches full. After the pump is withdrawn from the tank, fuel vapor continues to escape into the atmosphere in substantial quantities	Release toward end of refueling and after nozzle withdrawn
34	No vapor is seen escaping until the end of the refueling session, around 0:01:11, after which a substantial amount of fuel escapes into the atmosphere, continuing to escape even after the pump is withdrawn from the vehicle	Release toward end of refueling and after nozzle withdrawn
35	No vapor is seen escaping from the vehicle tank until the end of refueling. Vapors escape when the pump handle is partially withdrawn (0:01:12) and the tank is presumably topped off, and continue to escape even after the pump is fully withdrawn	Release toward end of refueling and after nozzle withdrawn
36	Although the pump is inserted into the vehicle from the beginning of the video, it appears that fuel is not dispensed until around 0:00:43 when the individual's hand squeezes the pump handle. As dispensing begins, large amounts of vapors can be seen escaping from the tank. Of note, the individual refueling does not fully insert the pump into the tank. Vapors escape nearly continuously throughout refueling, sometimes in large amounts. Toward the end of the session another large amount of vapor escapes, as the pump is pulled further out of the vehicle (0:01:55). Substantial amounts of vapor continue to escape until the end of refueling, including after the pump is fully withdrawn (0:02:49)	Near continuous vapor release
37	No vapor release observed	No vapor release
38	No vapor is observed until around 0:00:51, after which vapor is released nearly continuously. Vapor is observed escaping from the tank after the pump is withdrawn	Release toward end of refueling and after nozzle withdrawn
41	Some vapor is released at the beginning of the refueling session (0:00:14), but no more is observed until toward the end of refueling around (0:01:08). After this time, vapor is observed in substantial quantities until the pump is withdrawn (0:01:21), after which only minimal vapors are observed escaping	Release toward end of refueling
42	No vapors are observed until the very end of refueling, when the pump is withdrawn (0:01:59). Vapor continues to be released from the tank until it is capped	Release when nozzle withdrawn
43	No vapor release observed during refueling; a small amount of vapor may be released after pump is withdrawn (0:01:08)	Release when nozzle withdrawn
44	Poor video focus makes vapor observation difficult; however, ambient vapors appear to be present (upper right, 0:00:35, 0:00:40, 0:00:54)	Ambient vapors only
45	Substantial vapor release observed as cap is removed from tank, and continuously throughout refueling	Near continuous vapor release
46	No vapor release observed during refueling; a slight amount of release from pump observed as it was removed from tank (0:01:57)	Release when nozzle withdrawn
47	Slight amount of vapor release observed at start of refueling (0:00:03), and then again at end of refueling (0:00:24). Vapor continues to be released after pump removed	Release toward end of refueling and after nozzle withdrawn

vapors were released. Sorbent tubes attached to pumps, passive samplers, and real-time monitors are not able to do this because the amount of vapor measured is averaged over a time period, so it is challenging to determine when the vapor is released, or if it is released continuously.

Information about the timing of vapor releases is particularly useful because it can help researchers determine why vapors are being released. For example, ORVR systems with "liquid seals" are known to release some vapors at the end of refueling (13), because as the flow of gasoline into the vehicle tank decreases, the air gradient into the tank created by the moving gasoline decreases, allowing vapors to flow both into the tank and out of it (and thus into the atmosphere) (9). Release at the end of vehicle refueling was indeed one of our most common observations. However, vapor releases occurring in the middle of the refueling session, or throughout the session, both of which we observed in multiple refueling events, may suggest a breakdown in functioning of the ORVR system. These findings appear to be inconsistent with the ones by Tumbrink who did not observe measurable emissions during refueling (9). Ren and Hao in China did find measurable emissions throughout refueling, but at low levels, with vapor concentration increasing over time and ranging from 0 to 4.5 mg/m^3 (13). Emissions could be the result of a leak in part of the vehicle's fuel system, aging of the activation sites or oversaturation of the charcoal filter used in the ORVR, or a malfunctioning mechanical seal. It is also possible that that the pump nozzle itself is damaged, resulting in vapor release. In addition, Ren and Hao found that ambient temperature, fuel temperature, filling flow, and filling pipe diameter all have an impact on the time to liquid seal formation and on vapor emissions (13). Emissions were increased when either ambient or fuel temperature was higher (13). As our study was conducted at cold ambient temperatures (0.5–5°C), we expect that emissions during Spring, Summer, and Fall would be greater than what we observed.

Our study found an average refueling time of 86 s (1.43 min), similar to the 1.13 min found by Vainiotalo et al. (5) in Finland and less than that found by Egeghy et al. (7) in North Carolina (median of 3 min). These studies, and others, included various biomarkers and measures of exposure: internal dose (blood) (6), exhaled breath (7), and breathing zone air (5–7), all of which suggested individuals were exposed to benzene, a known human carcinogen, during refueling. As all studies were conducted before widespread adoption of ORVR and only at gas stations without Stage II vapor recovery, their results are likely not representative of the typical exposure today. Somewhat concerningly, however, our study suggests that despite extensive use of ORVR, individual exposures at similar magnitudes to those experienced before ORVR requirements were implemented may still occur—two of the three refueling

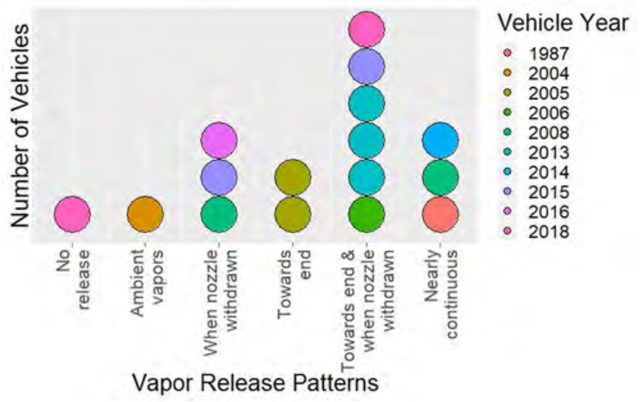

FIGURE 1 | Dotplot depicting the number of vehicles in each vapor release category (each dot represents one vehicle), with year delineated by color.

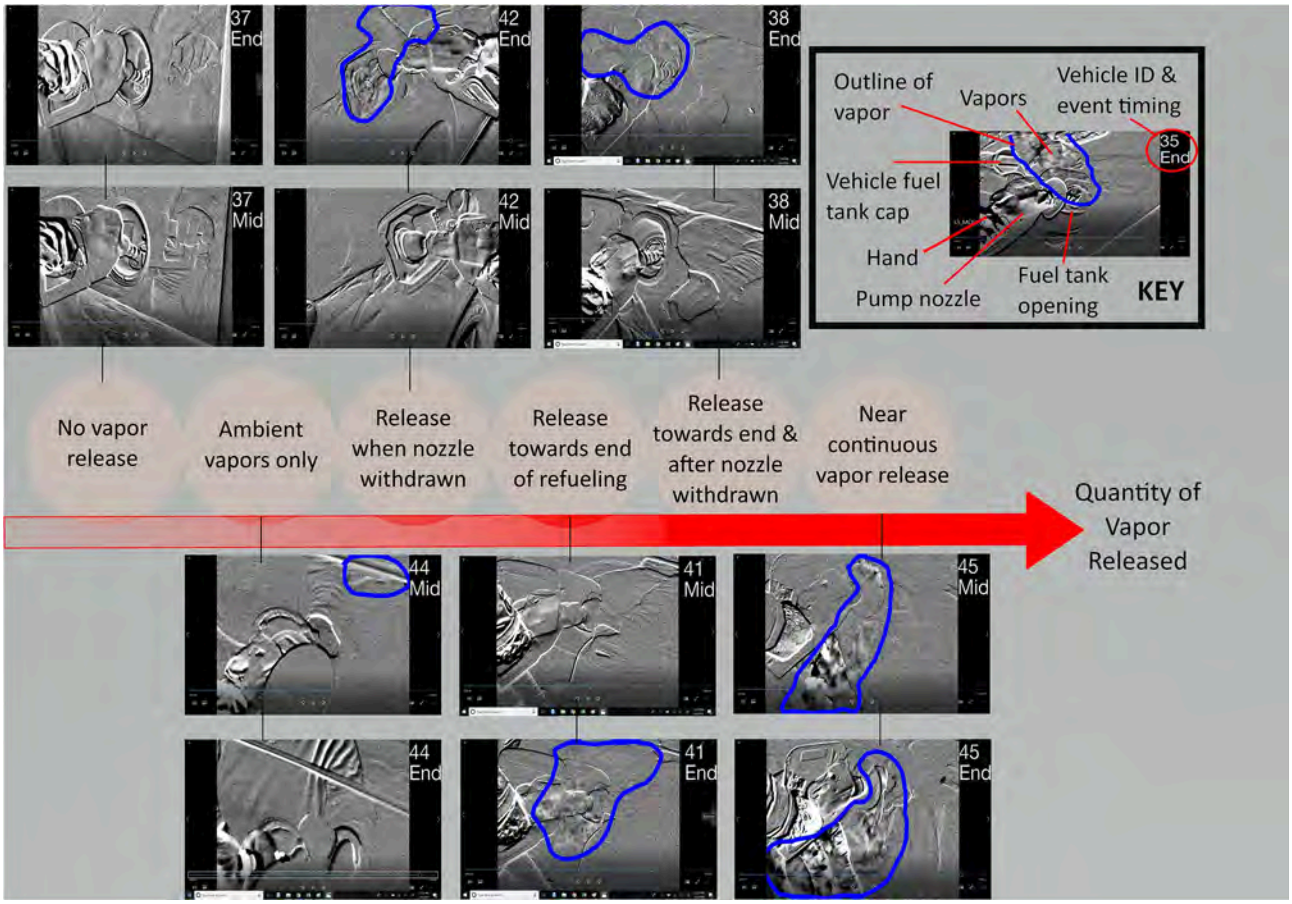

FIGURE 2 | Two sample frames for each of the 6 identified patterns of vapor release during refueling: one during the middle of the refueling session, and one at the end. Vehicle ID and an indicator for middle ("Mid") or end ("End") of the video are included in the upper right corner of each photo. Gasoline vapors, when present, are outlined by a blue line.

events categorized as "near continuous vapor release" happened in vehicles manufactured after the rollout of ORVR. Without Stage II vapor recovery, the population is not protected from emissions arising from the so-called legacy fleet without ORVR, vehicles with deteriorating ORVR, or motorcycles and boats, both of which do not have ORVR.

Of particular importance for public health and policy is the ability of ORVR systems to (1) reduce exposure to gasoline vapors during refueling to a safe level, and (2) continue to function at a high level over the lifetime of a vehicle. This is important for two reasons. First, volatile organic compounds (VOCs) released during refueling can chemically react in the atmosphere, contributing to ozone and other secondary pollutant formation, which can harm human health directly through cardiovascular pathways (14). ORVR systems are intended to reduce this potential, by preventing VOCs from escaping into the atmosphere where they can react with other species. Second, as previously discussed, exposure to primary VOCs, such as those in gasoline can also negatively impact health directly, from exposure during vehicle refueling. However, limited work has been conducted to test the assumption that ORVR reduces exposure to a "safe" level during vehicle refueling. In fact, it is unclear what a "safe" level of exposure to gasoline vapors is, particularly as there is not a standardized formula for gasoline.

Numerous studies have been conducted (15, 16) to characterize the potential harms of gasoline with specific formulas or additives, but these reports typically compare different formulas of gasoline rather than comparing exposure to no exposure. Evidence suggests that while exposure during refueling is likely, health effects from gasoline at infrequent low-levels may be small, although individual components are carcinogenic (15, 16). Conversely, evidence from occupational studies has shown that individuals chronically exposed to lower levels of gasoline vapors, for example gas station attendants, are at higher risk for certain cancers (17, 18). Despite this evidence, we do not fully understand what risk gasoline vapors pose to the general public during typical vehicle refueling, or the cumulative impact of such exposure over an individual's lifetime, particularly in today's regulatory environment. Our findings highlight, in a visually compelling manner, that individuals can be exposed to substantial amounts of gasoline vapors during refueling, even in a vehicle fleet saturated with ORVR.

Future studies are vitally needed to determine the risk to individuals during typical refueling sessions in a vehicle fleet saturated with ORVR, especially because exposure to gasoline is ubiquitous and occurs throughout the lifetime. We recommend comprehensive exposure assessments that estimate exposure, internal dose, and health effects, as well as real-time monitoring of volatile organic compounds, potentially using a portable SHED (19) deployed at a gas station and paired with an infrared camera optimized for gas imaging. In addition, we recommend future work to develop an algorithm for estimating the amount or concentration of vapors shown in video from an infrared camera, to provide a better understanding of the concentration of vapors dispersing around a station.

This pilot study has several limitations. First, a convenience sample of stations and vehicles were used, and thus may not be representative of the true vehicle fleet in NYC. However, ORVR saturation in our sample was fairly close to an estimate for all registered vehicles in New York State and City (94 vs. 81%). It is additionally reassuring that both these estimates are above the EPA estimate of 71% for ORVR saturation in the older 2012 US fleet (8) and that the saturation in our convenience sample is above New York State's modeled estimate of 85% or greater for the older 2013 fleet (20). The median manufacturing year of our sample was consistent with that for New York State and City's registered vehicles (median = 2013). Second, the small sample size does not provide ample power for statistical tests. Third, vehicle make, model, and age were estimated by researchers and therefore there is potential for misclassification. Finally, real-time estimates of VOC concentrations were not obtained.

CONCLUSIONS

In an ORVR saturated vehicle fleet, use of an infrared camera optimized for VOC imaging allowed for the identification of vapor sources, viewing vapor trajectory and dispersion, and identifying the timing of vapor release during refueling. In this pilot study, 14 out of 16 observed refueling events resulted in vapor emissions, with severity varying substantially by vehicle make/model and age. A full exposure-assessment incorporating infrared cameras, quantitative monitors, and biologic samples is needed to understand exposure to and health effects of fuel vapor at gas stations, in an ORVR saturated vehicle fleet.

DATA AVAILABILITY STATEMENT

All datasets generated for this study are included in the article/**Supplementary Material**.

AUTHOR CONTRIBUTIONS

MH and JS conceptualized the study and completed data collection. JS wrote the first manuscript draft and completed initial data analysis. MH supervised and reviewed all the data analysis and edited the manuscript. All authors agree to be accountable for the content of this work.

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SUPPLEMENTARY MATERIAL

The Supplementary Material for this article can be found online at: https://www.frontiersin.org/articles/10.3389/fpubh.2020.00018/full#supplementary-material

REFERENCES

1. Hilpert M, Mora BA, Ni J, Rule AM, Nachman KE. Hydrocarbon release during fuel storage and transfer at gas stations: environmental and health effects. *J Curr Environ Health Rep.* (2015) 2:412–22. doi: 10.1007/s40572-015-0074-8
2. Hilpert M, Rule AM, Adria-Mora B, Tiberi T. Vent pipe emissions from storage tanks at gas stations: implications for setback distances. *Sci Total Environ.* (2019) 650:2239–50. doi: 10.1016/j.scitotenv.2018.09.303
3. ATSDR. *Toxicological Profile for Benzene. Agency for Toxic Substances and Disease Registry*. Atlanta, GA: US Department of Health and Human Services (2007).
4. Abou-ElWafa HS, Albadry AA, El-Gilany AH, Bazeed FB. Some biochemical and hematological parameters among petrol station attendants: a comparative study. *Biomed Res Int*. (2015) 2015:418724. doi: 10.1155/2015/418724
5. Vainiotalo S, Peltonen Y, Ruonakangas A, Pfäffli P. Customer exposure to MTBE, TAME, C6 alkyl methyl ethers, and benzene during gasoline refueling. *Environ Health Perspect.* (1999) 107:133–40. doi: 10.1289/ehp.99107133
6. Backer LC, Egeland GM, Ashley DL, Lawryk NJ, Weisel CP, White MC, et al. Exposure to regular gasoline and ethanol oxyfuel during refueling in Alaska. *Environ Health Perspect.* (1997) 105:850–5. doi: 10.1289/ehp.97105850
7. Egeghy PP, Tornero-Velez R, Rappaport SM. Environmental and biological monitoring of benzene during self-service automobile refueling. *Environ Health Perspect.* (2000) 108:1195–202. doi: 10.1289/ehp.001081195
8. EPA. 40 CFR Part 51: Air Quality: Widespread Use for Onboard Refueling Vapor Recovery and Stage II Waiver. *Environmental Protection Agency. Federal Register*. (2012) 77:28772–82.
9. Tumbrink, M. *Filtersysteme im Automobil: Innovative Lösungsansätze für die Automobilindustrie [Filtration Systems in the Car: Innovative Solution Approaches for Car* Manufacturers]: Haus de Technik–Fachbuchreihe (Tübingen) (2002).
10. Schifter I, Díaz L, Rodríguez R, González-Macías C. The contribution of evaporative emissions from gasoline vehicles to the volatiel organic compound inventory in Mexico City. *Environ Monit Assess.* (2014) 186:3969–83. doi: 10.1007/s10661-014-3672-2
11. R Core Team. *R: A Language and Environment for Statistical Computing.* Vienna: R Foundation for Statistical Computing (2018).
12. Wickham H. *Tidyverse: Easily Install and Load the 'Tidyverse'*. R package version 1.2.1 (2017).
13. He R, Ding H. Refueling experiment of on-board refueling vapor recovery. *China J Highw Transp.* (2017) 30:142–50.
14. Zhao R, Chen S, Wang W, Huang J, Wang K, Liu L, et al. The impact of short-term exposure to air pollutants on the onset of out-of-hospital cardiac arrest: a systematic review and meta-analysis. *Int J Cardiol.* (2017) 226:110–7. doi: 10.1016/j.ijcard.2016.10.053
15. ATSDR. *Toxicological Profile for Gasoline*. Agency for Toxic Substances and Disease Registry. Atlanta, GA: US Department of Health and Human Services (1995).
16. NSCAUMATC. *Evaluation of the Health Effects From Exposure to Gasoline and Gasoline Vapors*. Northeast States for Coordinated Air Use Management Air Toxics Committee (1989).
17. Morton W, Marjanovic D. Leukemia incidence by occupation in the Portland-Vancouver metropolitan area. *Am J Ind Med.* (1984) 6:185–205. doi: 10.1002/ajim.4700060304
18. Schwartz E. Proportionate mortality ratio analysis of automobile mechanics and gasoline service station workers in New Hampshire. *Am J Ind Med.* (1987) 12:91–9. doi: 10.1002/ajim.4700120110
19. Eastern Research Group. *Denver Summer 2008 Pilot Study at Lipan Street Station–Report Version 5*. Assessment and Standards Division, Office of Transportation and Air Quality and United States Environmental Protection Agency (2008).
20. NYDEC. *Stage II Vapor Collection System Enforcement Discretion Directive*. New York, NY: State Department of Environmental Conservation (2011). Available online at: https://www.dec.ny.gov/regulations/74990.html (accessed November 30, 2019).

Conflict of Interest: The authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

2021 Study
Benzene emissions from gas station clusters: a new framework for estimating lifetime cancer risk

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RESEARCH ARTICLE

Benzene emissions from gas station clusters: a new framework for estimating lifetime cancer risk

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Abstract

Purpose During gas station operation, unburned fuel can be released to the environment through distribution, delivery, and storage. Due to the toxicity of fuel compounds, setback distances have been implemented to protect the general population. However, these distances treat gasoline sales volume as a categorical variable and only account for the presence of a single gas station and not clusters, which frequently occur. This paper introduces a framework for recommending setback distances for gas station clusters based on estimated lifetime cancer risk from benzene exposure.

Methods Using the air quality dispersion model AERMOD, we simulated levels of benzene released to the atmosphere from single and clusters of generic gas stations and the associated lifetime cancer risk under meteorological conditions representative of Albany, New York.

Results Cancer risk as a function of distance from gas station(s) and as a continuous function of total sales volume can be estimated from an equation we developed. We found that clusters of gas stations have increased cancer risk compared to a single station because of cumulative emissions from the individual gas stations. For instance, the cancer risk at 40 m for four gas stations each dispensing 1 million gal/year is 9.84×10^{-6} compared to 2.45×10^{-6} for one gas station.

Conclusion The framework we developed for estimating cancer risk from gas station(s) could be adopted by regulatory agencies to make setback distances a function of sales volume and the number of gas stations in a cluster, rather than on a sales volume category.

Keywords Gas station clusters · Cancer risk · Benzene · VOC emissions · Air pollution modeling

Introduction

In 2016, about 143 billion gallons of gasoline were dispensed at United States (US) gas stations [1]. This is equivalent to an average consumption of 442 gal of gasoline per person [2]. During the operation of a gas station, unburned fuel is released from multiple sources, including spills, leaky pipes, leaky dispenser hoses, leaks in underground storage tanks, and underground storage tank venting [3–6]. All of these sources of exposures can contribute to negative health effects due to the toxicity of chemicals in unburned fuel.

Gasoline contains the BTEX group, consisting of benzene, toluene, ethylbenzene and xylenes, all of which are toxic [7–9]. Within the BTEX group, benzene is the sole chemical classified as a human carcinogen [10]; it is a causal agent of leukemia and is associated with non-Hodgkin's lymphoma and multiple myeloma [7, 11]. While the general population experiences low exposure to benzene at gas stations when dispensing gasoline, at-risk populations include those who are occupationally exposed, people that live near gas stations, and children in schools near stations [12–16]. People living near gas stations can be exposed to chemicals from the stations even inside their homes, as modeled by Hicklin et al. [17] in Malta and measured by Barros et al. [18] in Portugal. Additionally, studies suggest that there may be a risk of childhood leukemia associated with living close to gas stations [19–22]. Yet another study concluded that the lifetime cancer risk at and around selected gas stations in Iran exceeded values proposed by the US Environmental Protection Agency (EPA) [23].

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As cancer risk due to toxic evaporative emissions from a gas station is a function of distance from the gas station [24, 25], regulations in the form of setback distances have been put in place to protect people. In the US, different states have different guidelines for setback distances, and even within states different counties may set their own guidelines. Based on estimations of lifetime cancer risk, the California Air Resources Board (CARB) recommends that new sensitive land uses (such as schools and daycares) should not be sited within 300 ft (91 m) of a large gasoline dispensing facility, where large is defined as having a sales volume of at least 3.6 million gallons per year [26, 27]. On the other hand, a county council in the US state of Maryland approved a zoning amendment that requires gas stations that pump more than 3.6 million gallons of gas per year to be 500 ft. from public and private schools, parks, playgrounds, recreational areas, homes, and environmentally sensitive areas [28]. In these examples, sales volume is treated as a categorical value, which results in a loss of precision and assumes the relationship between exposure and cancer risk is the same for all sales volumes in a given category. Moreover, we are unaware of any setback distances that account for the presence of gas station clusters. To improve regulations around setback distances for gas stations, the effects of sales volume and number of gas stations in a cluster on cancer risk due to fuel spills and evaporative fuel losses should be examined.

To inform recommendations for setback distances from gas stations, we performed air dispersion modeling to obtain the spatial distribution of lifetime cancer risk due to benzene emissions from single gas stations and clusters, making assumptions about evaporative emission rates from gas stations and meteorological conditions that are representative of Albany, New York. The main objectives of this paper are to (1) examine how lifetime cancer risk due to evaporative benzene releases depends on sales volume and the number of gas stations in a cluster and (2) to introduce a framework for recommending setback distances between gas stations and adjacent sensitive land uses based on estimated lifetime cancer risk from benzene exposure. Unlike previous work [24, 26], this framework treats sales volume as a continuous variable, accounts for cumulative emissions from gas station clusters, and allows calculating cancer risk by evaluating an equation instead of reading it from a plot.

Methods

Meteorological data

We used three years of hourly meteorological data (2015–2017) for Albany, New York in the US. A location in the state of New York was chosen, because we wanted our work to be relevant to a local community. We chose Albany over New York City, however, because New York City has generally much taller buildings, which would need to be accounted for in pollutant dispersion simulations, something that is typically avoided when modeling health effects from generic gas stations [24, 29]. The surface air data were obtained from the National Climatic Data Center for the Albany International Airport, and the upper air data were obtained from the NOAA/ESRL Radiosonde Database for Albany, NY [30]. Descriptive statistics of the meteorological data were obtained with the ‘openair’ package in R 3.5.1 and are shown in Supplementary Table 1, and the wind rose is shown in Supplementary Fig. 1.

Generic gas station modeling

We assumed that gas station clusters consist of up to $N_{st} = 4$ gas stations located on the four corners of an intersection (even though other configurations are possible). Figure 1 depicts the four-gas station configuration. Each of the four gas stations is assumed to have four pump islands, from which fuel can be dispensed from both sides. At each gas station, the underground storage tank vent pipe is assumed to be in the center of the gas station, even though they are often located at the fence line or on the walls of convenience stores or auto repair shops, which are often part of a gas station. For configurations with more than one gas station, the origin of the modeling domain is the center of the intersection. For a single gas station configuration, the origin is the center of that gas station. This assumption was made for better comparability between the cancer-distance relationships for single and clusters of gas stations. Figure 2 depicts three-, two-, and one-gas station configurations. In Figs. 1 and 2, the origin is indicated by a red plus sign.

Air dispersion modeling

To model the dispersion of benzene vapors released into the atmospheric environment through evaporative losses from gas station clusters, we used AERMOD and AERMET, regulatory software developed by the US EPA [31, 32]. The AERMOD software models hourly levels of air pollutants in gas or particulate phase in the atmospheric boundary layer based on a steady-state plume approach that accounts for meteorological, topographic, surface roughness and emission source information [33]. AERMOD was compared to 16 tracer release field studies, and with few exceptions was found to have superior model performance when compared to other models tested [34]. The AERMET software was used to pre-process meteorological data for input into AERMOD. Benzene levels for subsequent cancer risk estimations were modeled on two-dimensional polar grids at different radial distances r_j (from 0 to 200 m in 20-m steps) and different angles φ_i (from 10° to 360° in 10° steps). Benzene levels were simulated at a 1-h temporal resolution at three elevations, $z = 0$, 2 and 4 m,

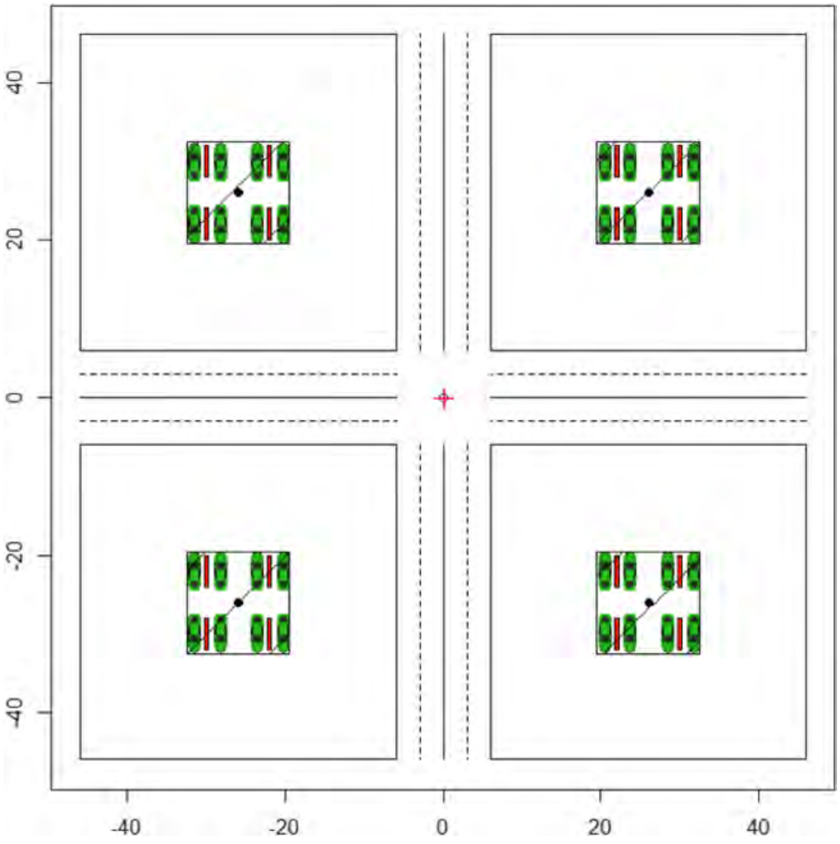

Fig. 1 Generic gas station cluster with one gas station on each corner of an intersection (drawn to scale except for enlarged vent pipes). Each gas station can accommodate two vehicles (green) per pump island (red) and has one vent pipe in the center (black dot). Diagonal lines indicate gas station canopies. Axes labels indicate distance in meters. The red "+" represents the origin of the modeling domain

representative of the ground-level, the breathing zone, and a second-floor level residence, respectively. We configured AERMOD to calculate the 3-year temporal averages of the hourly time series of the simulated concentration fields. For visualizing the simulated 3-year average benzene levels, much finer numerical grids that were particularly well resolved around the benzene sources were used to create contour plots of benzene levels using Matlab™ R2017b version.

Emission modeling

Emissions of unburned gasoline from gas stations depend on installed pollution prevention technologies. We assumed presence of pollution technology that is representative or will become representative for most US states (with the notable exception of California). Based on these assumptions, we simulated California Air Pollution Control Officers Association's (CAPCOA) Scenario 5B ("Phase I" with vent valves, underground storage tank) [24].

Specifically, we assumed presence of Stage I vapor recovery, which reduces the amount of fuel vapors that would be pushed into the atmosphere during the refueling of underground storage tanks by the rising fuel levels in the tanks by directing these vapors into tanks on the delivering tanker truck. We assumed the absence of Stage II vapor recovery, because EPA has recently allowed states not to require Stage

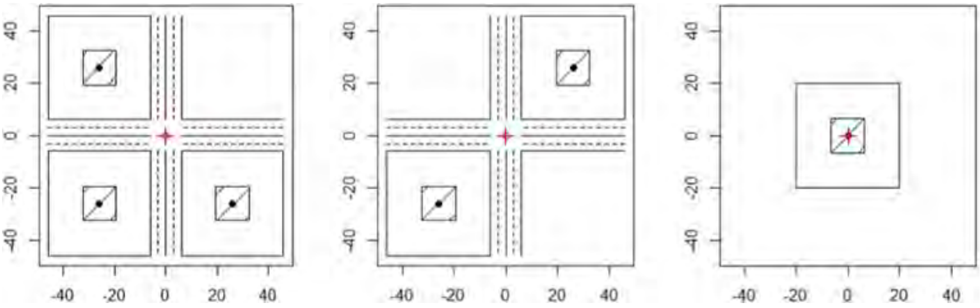

Fig. 2 Simplified depictions of generic gas station clusters consisting of three, two and one gas stations (drawn to scale except for enlarged vent pipes). Each station has one vent pipe in the center (black dot). Diagonal lines indicate gas station canopies. The red "+" represents the origin of the modeling domain. Axes labels indicate distance in meters

II systems if widespread use of Onboard Refueling Vapor Recovery (ORVR) is given [35].

The refueling emission factor we used accounts for the fact that not all vehicles (e.g., legacy fleet, motorcycles) are equipped with ORVR. We assumed an ORVR penetration rate (PR) of 93.2% which represents the percentage of gasoline dispensed to ORVR-equipped vehicles that has been estimated for the US for the year of 2019 [35]. We assumed 95% for the efficiency of ORVR [35], i.e., refueling losses from ORVR-equipped vehicles are 5% of the losses from non-ORVR equipped vehicles, which is 8.4 lbs./kgal. Thus the refueling loss is given by: $[(1 - \mathrm{PR}) + 0.05 \times \mathrm{PR}] \times 8.4$ lbs./kgal = 0.96 lbs./kgal. Table 1 summarizes the emission losses we assumed.

To convert gasoline losses into benzene emission rates, we made assumptions about fuel composition. We assumed that current US liquid gasoline (except in California) contains about 1% of benzene by volume [36]. Like CAPCOA [24] and Hilpert et al. [29], we assumed a mass fraction of benzene in the ullage/headspace of the underground storage tank of 0.003 (by weight benzene in vapors) [29].

Emission factor values were used to calculate the parameter values for the AERMOD input file. For a 1-gas station configuration, we defined a total of nine sources: one vent pipe, four refueling and hose permeation loss sources (combined for each pump island), and four spillage loss sources (one for each pump island). We think of a gas station as having point and volume sources. Refueling, hose permeation and spillage losses were modeled as volume sources because they do not occur at fixed locations since the locations of different customer vehicles vary even if the same pump is used for refueling. For all volume sources, we assumed an initial lateral dimension of 3.02 m (stated as SYINIT in Table 2) and initial vertical dimension of 1.86 m (stated as SZINIT in Table 2), which are based on previous modeling assumptions for gas stations. The release height (stated as HS in Table 2) of the spillage losses was assumed to be at the ground-level elevation, because spilled droplets fall to the ground, where most of the evaporation takes place, while the release height for refueling and hose permeation was assumed to be 1 m. Vent pipe losses were modeled as point sources because underground storage tank vent pipes extend up above the surface of the pavement behaving more like a chimney emission rather than a volume emission. For vent pipe sources, the altitude from the ground was assumed to be 4 m (stated as HS in Table 2). For each gas station, all emission sources were assumed to be located at its center. Table 1 describes the source parameters.

Table 1 Emission factors

Type	Loss (lbs/kgal)*
Loading	0.084
Breathing	0.21
Refueling for 0% ORVR penetration	8.4
Refueling for assumed 93.2% ORVR penetration	0.96
Spillage	0.61
Hose permeation	0.062

*In the US, regulatory agencies typically express emission losses in units of lbs./kgal, i.e., pounds of gasoline emitted/lost per 1000 gal of gasoline dispensed

Note that 1 lbs./kgal = 0.1198 kg/m^3 *

Table 2 shows selected input parameters for AERMOD simulations. Note that the SYINIT (initial lateral dimension of the volume source [SYINIT]) of 3.02 m was obtained by dividing the canopy width (13 m) by 4.3, a constant, which is based on previously developed modeling assumptions for gas stations [24]. The vent pipe exit velocity was calculated from the sales volume SV_{single}, the assumed inside diameter of the vent pipe (2 in = 5.1 cm), and the loading and breathing emission factors from Table 1.

Our single generic gas station was assumed to have a sales volume SV_{single} = 1,000,000 gal/yr. Even though the dependence of stack exit velocity on sales volume causes simulated benzene concentration fields to depend non-linearly on sales volume, this non-linearity is negligible. A comparison between the concentration field simulated for the actual stack exit velocity and the field for a hypothetical stack exit velocity of zero showed that concentrations differed by no more than 0.3% on the numerical grid points. Therefore, concentration fields for other sales volumes can be estimated from the simulations for SV_{single} = 1,000,000 gal/yr by assuming a linear scaling law between the benzene concentration field for SV_{single} = 1,000,000 gal/yr and the actual sales volume. Finally we assumed no buildings to be present and flat terrain.

Cancer risk modeling

Cancer risk (CR) from inhalation exposure to benzene was modeled using the concept of Inhalation Unit Risk (IUR), which is an estimate of the increased cancer risk from inhalation exposure to a concentration of 1 μg/m^3 for a lifetime [37]. EPA estimates IUR to be between 2.2×10^{-6} per μg/m^3 and 7.8×10^{-6} per μg/m^3 [37]. Lifetime cancer risk from benzene was calculated according to EPA guidelines for inhalation risk assessment [37]. Thus, cancer risk at each point of the numerical grid can be calculated as follows:

$$CR = IUR\ x\ EC \tag{1}$$

where EC (μg/m^3) is the spatially variable exposure concentration or intake. The intake is calculated from EC = (CA x ET x EF x ED) / AT where CA (μg/m^3) is the benzene concentration modeled at each grid point and averaged over the entire

Table 2 Selected input parameters for AERMOD simulations

Description	Emission rate QS (g/s)	Release height HS (m)	Stack exit emperature TS (Kelvin)	Exit velocity VS (m/s)	Stack diameter DS (cm)	Initial lateral dimension of volume SYINIT (m)	Initial vertical dimension of volume SZINIT (m)
Hose permeation losses and refueling losses combined	0.0001567	1.0	N/A	N/A	N/A	3.02	1.86
Spillage losses	0.0003159	0.0	N/A	N/A	N/A	3.02	1.86
Vent pipe loading and breathing losses combined	0.0001522	4.0	290	0.001236	5.1	N/A	N/A

Abbreviations: *N/A* not applicable

simulation period (3 years), ET (hours per day) is the exposure time, EF (days per year) is the exposure frequency, ED (years) is the exposure duration, and AT (hours) is the average time per exposure period. We chose EPA's upper bound for IUR which would be appropriate for a sensitive land use and exposure parameters indicative of constant presence e.g. children in a boarding school or residents in a nursing home: ET = 24 h/day, EF = 350 days/year (7 days/week × 52 weeks/year), ED = 70 years (lifetime cancer risk), and AT = 613,200 h (70 years × 365 days/year × 24 h/day) [37]. We therefore calculated the lifetime cancer risk as follows: $CR = 7.8 \times 10^{-6}$ $(\mu g/m^3)^{-1}$ x EC.

To facilitate estimation of cancer risk of the various gas station clusters as a function of distance r from the gas station and the total sales volume $SV_{tot} = N_{st}$ SV_{single} where N_{st} represents the number of gas stations, we fitted a simple mathematical model to the spatial distribution of modeled cancer risk. This model condenses the concentrations simulated on the two-dimensional polar grid onto a one-dimensional grid where concentration is expressed as a function of distance r from the origin of the model domain: $\langle \mathrm{CR} \rangle \left(r_j\right) = \frac{1}{N}\sum_{i=1}^{N} \mathrm{CR} \left(r_j, \varphi_i\right)$ where $N = 36$ is the number of discrete angles used in the numerical grid. We assumed that the dependence of cancer risk $\langle \mathrm{CR} \rangle$ on distance r is described by an exponentially decaying function according to the following equation:

$$\log_{10}\left(\langle \mathrm{CR} \rangle \frac{10^6 \mathrm{gal/yr}}{N_{st}\ \ \mathrm{SV}_{single}}\right) = a + br \tag{2}$$

As shown in Section A in Supplementary Material, Eq. (2) is consistent with empirical Gaussian plume models [38].

Also note that the cancer risk scales linearly with sales volume SV_{single}, consistent with the AERMOD simulations, which yields concentration fields that scale linearly with benzene source terms. Therefore, regressions coefficients a and b do not depend on which value of SV_{single} is chosen in the simulations. We also assumed cancer risk to depend linearly on N_{st}; however, a and b can be expected to show some dependence on N_{st} because benzene levels at any grid point do not scale exactly linearly with N_{st} as the gas stations in the cluster have typically different distances to a grid point. We therefore did not only determine a and b by fitting simultaneously the modeled spatial distributions of cancer risk for all gas station configurations to Eq. (2), but we also determined for each gas station configuration alone a and b and then used one-way ANOVA to examine potential differences between regression coefficients among the four gas station configurations (significance level of 0.05). The goodness of fit was evaluated with the R^2 value. In the regressions, we excluded the first two data points for distances 0 and 20 m from the regressions, because inclusion would have increased the variance of the regression too much since for these distances normalized cancer risks were too different across the four cluster types.

Cancer risk modeling and analyses were completed using R 3.5.1 (R Foundation for Statistical Computing, Vienna, Austria).

Results

Air pollution modeling

Figure 3 shows simulated atmospheric benzene levels arising from the gas station cluster, which contains four gas stations, for three different elevations. Generally, benzene levels decrease with distance from each gas station until the influence of one of the other three gas stations is felt; then levels may increase again. Further away from the intersection and the entire gas station cluster, benzene levels generally decrease. Benzene level fields do not exhibit any symmetry, and levels are not constant along circles of radius r around the center of the modeling domain.

Close to the intersection (< 60 m), benzene levels depend substantially on elevation. At the 4-m elevation around the vent pipes, the only modeled point sources of benzene, concentrations tend to be highest. Further away from the intersection (>80 m), benzene levels do not depend much on elevation.

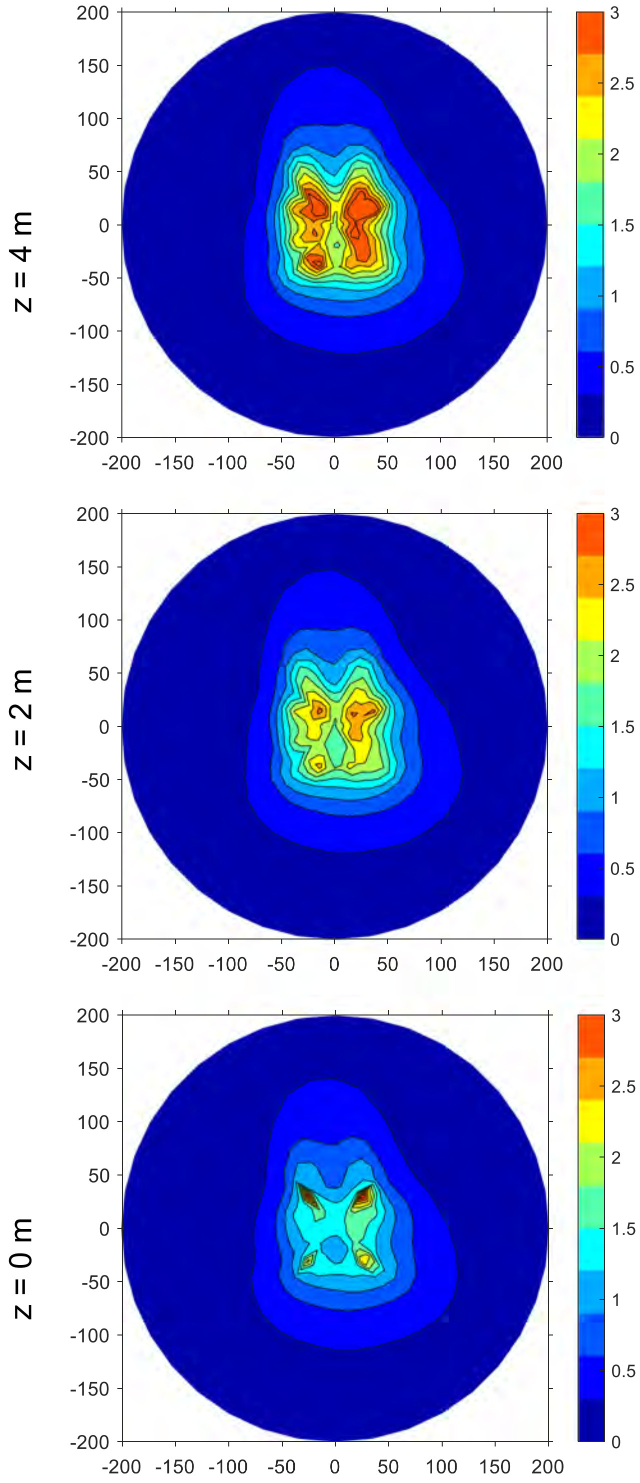

Fig. 3 Modeled atmospheric benzene levels (3-year average) due to emissions from four-gas station configuration shown at 3 elevations: 0 (bottom panel), 2 (middle panel), and 4 m (top panel). Abscissa and ordinate labels indicate distance in meters. Color bar indicates benzene concentration in µg/m^3

Figure 4 shows simulated atmospheric benzene levels in the breathing zone that arise from the four different gas stations clusters. Benzene levels clearly depend substantially on the number of gas stations present. Moreover, the spatially dependent concentration fields for more than one gas station cannot simply be obtained by multiplying the concentration field for one gas station by the number of gas stations in the cluster.

Cancer risk modeling

Figure 5 shows boxplots of the log-transformed cancer risk normalized by total sales volume (left-hand-side of Eq. (2)) as a function of distance from the origin of the modeling domain. For distances ≥40 m, median normalized cancer risks are roughly the same for the four configurations. For distances <40 m (0 and 20 m), however, these risks differ substantially between configurations. Specifically, the single gas station exhibits different patterns, with cancer risk monotonically decreasing with distance; whereas for the configurations with more than one gas station cancer risk is greatest at a distance of 20 m. The heights of the box plots (interquartile range) in Fig. 5 also illustrate that cancer risk for a given distance and gas station configuration can vary by more than a factor of 10 depending on the angle φ_i.

Figure 6 shows the linear regressions for the log-transformed cancer risk medians, normalized by total sales volume, for the four different gas station configurations. Results from the regression analyses are summarized in Table 3. For all regressions, R^2 values are >0.96, and the F statistics are statistically significant ($p < 0.05$). In addition, all intercept and regression coefficients are statistically significant ($p < 0.05$), meaning distance and lifetime cancer risk are significantly associated. One-way ANOVA showed that regression coefficients a and b are not different across the four gas station configurations. At the same time, confidence intervals (CIs) between coefficients across gas station configurations overlapped. CIs of the regression coefficients that account for the data of all gas station configurations together overlap with the CIs from the four individual regressions.

Summary and discussion

Spatial dependence of benzene levels

We for the first time presented simulations for the cumulative effects of several gas stations on atmospheric benzene levels. As previously established, benzene levels depend substantially on distance from gas station [12–15, 25]; however, similar to Hilpert et al. [29], we also found that elevation is a determining factor [29]. Benzene levels on the ground surface (0-m elevation) and in the breathing zone (2-m elevation) are similar to each other (Fig. 3), because at lower elevations benzene levels arise from volume and surface forces and are not affected much by vent pipe emissions. Close to a gas station (<

Fig. 4 Modeled atmospheric benzene levels (3-year average) due to emissions from 4, 3, 2, and 1 gas station configuration at an elevation of 2 m. Abscissa and ordinate labels indicate distance in meters. Color bar indicates benzene concentration in μg/m^3

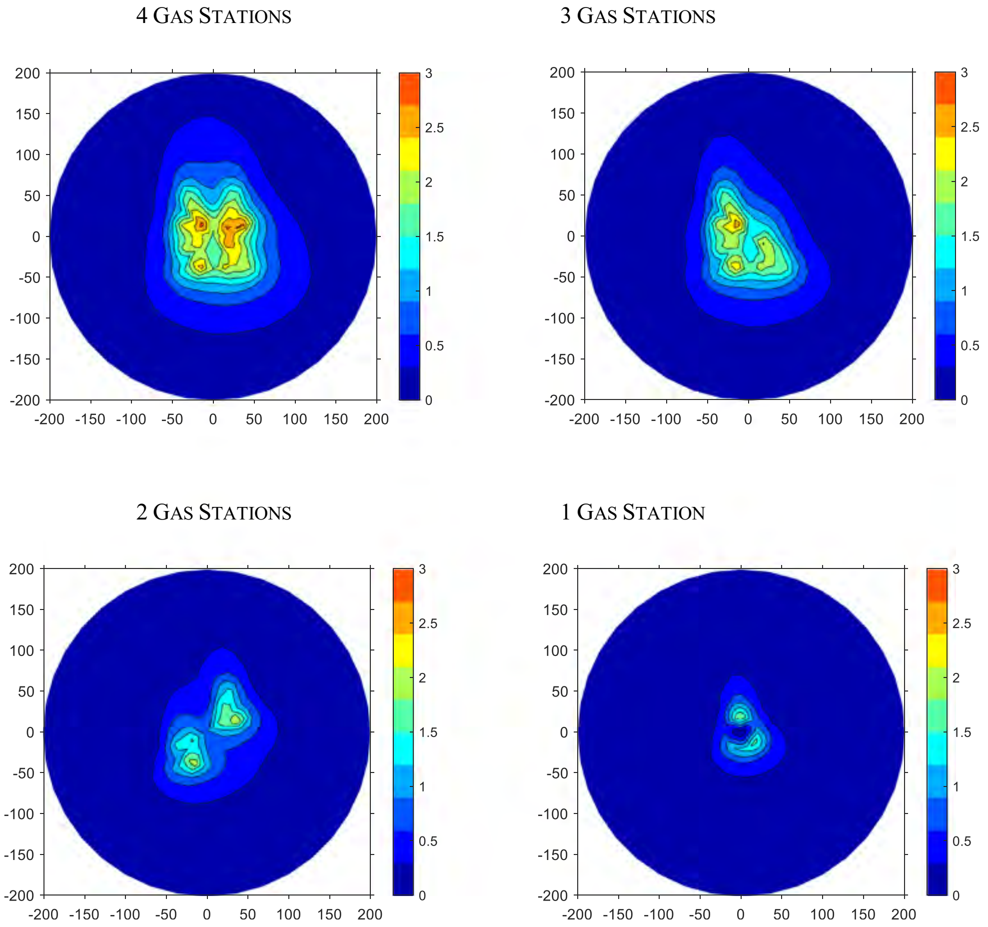

Fig. 5 Lifetime cancer risk <CR> normalized by total sales volume and then log-transformed for four different gas station clusters consisting of 1, 2, 3 and 4 gas stations by distance r from the origin of the model domain. Box plots indicate the variation of cancer risk at distance r due to its dependence on the angle φ_i at the $z = 4$ m elevation

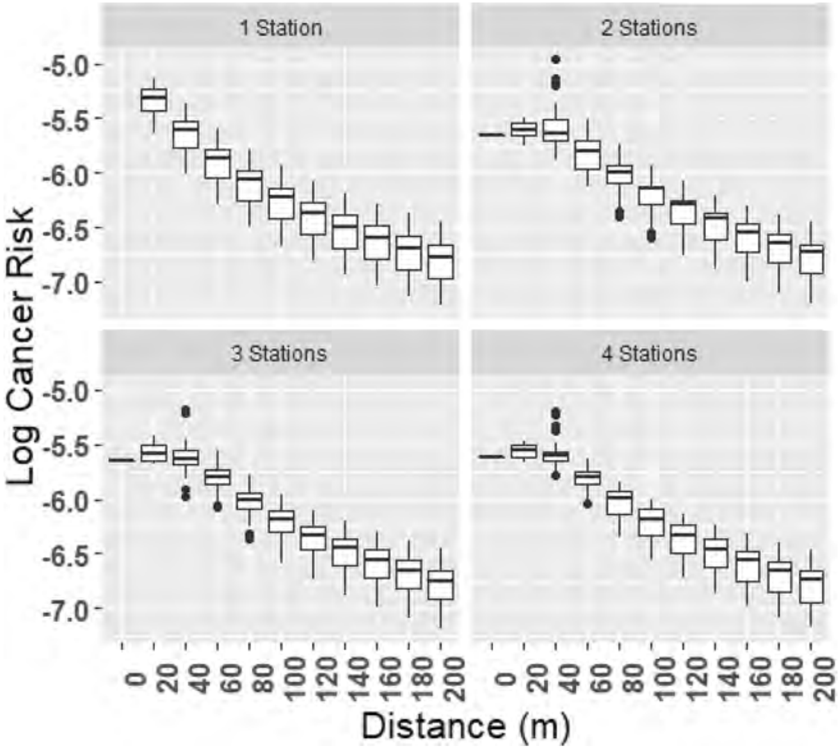

Table 3 Summary of linear regression for medians of lifetime cancer risk according to Eq. (2)

# Gas Stations	All	4	3	2	1
Intercept *a* [95% CI]	−5.50 [−5.55, −5.45]	−5.40 [−5.53, −5.28]	−5.42 [−5.53, −5.30]	−5.41 [−5.51, −5.32]	−5.45 [−5.61, −5.30]
Distance coefficient *b* (1/km)* [95% CI]	−6.49 [−6.91, −6.07]	−7.12 [−8.10, −6.15]	−7.04 [−7.92, −6.15]	−6.92 [−7.62, −6.22]	−7.03 [−8.19, −5.87]
R-squared	0.96	0.98	0.98	0.99	0.97
Cancer Risk at 40 m	N/A	9.84×10^{-6}	6.94×10^{-6}	4.66×10^{-6}	2.45×10^{-6}

*All intercepts and distance coefficients are statistically significant ($p < 0.05$)

40 m), benzene hot spots are present at a 4-m elevation where the vent pipes of the fuel storage tanks were assumed to release fuel vapors to the atmospheric environment, potentially putting residents at the 2nd floor level at risk. Further away from the center of the modeling domain (about >80 m), concentration fields do not depend much on elevation, as evidenced by the almost identical contour lines for benzene levels. This is because of vertical mixing of the benzene vapors due to atmospheric dispersion. Additionally, for quality assurance, we conducted a simulation for a single gas station where stack velocity is zero and compared the benzene concentration levels to our results (which use a stack velocity of 0.0012). We found that the percent difference for benzene concentration between the two simulations was close to zero.

Cancer risk as a function of sales volume and number of gas stations

We performed for the first time analyses that not only allow estimating cancer risk of a single gas station as a function of sales volume but also the risk from multiple gas stations in a cluster. In contrast, previous reports presented examples of cancer risk as a function of distance r only from a single gas station in the form of plots for a given sales volume SV, with no guidance about how to estimate cancer risk for a different sales volume. See, for example, Appendix E in CAPCOA [24] which presents cancer risks for gas stations dispensing 1 million gal/yr or Figs. 1, 2, 3, 4, 5 and 6 in CalEPA/CARB [26] for a gas station dispensing 3.6 million gal/yr [24, 26]. Our plots and Eq. (2), both of which normalize cancer risk by sales volume, respond to this need. For instance, one can now easily answer the question: what is the lifetime cancer risk <CR> of a single gas station dispensing 10 million gal/yr at a distance $r = 150$ m? We can read from the red line in Fig. 6, that $\log_{10}(\ldots) \sim -6.5$ and therefore $10^{-6.5} = (\mathrm{CR})\frac{10^6\,\mathrm{gal/yr}}{N_{st}\ \mathrm{SV}_{single}}$. Since $N_{st} = 1$ and $\mathrm{SV}_{\mathrm{single}} = 10^7$ gal/yr, the cancer risk is $\langle \mathrm{CR} \rangle = 10^{-5.5}$ which is 3 in a million.

Directional dependence of cancer risk

At a single location (specified by distance r and angle φ_i), substantial differences between the cancer risk inferred from Eq. (2) and the risk calculated from Eq. (1) using the AERMOD benzene concentration at that location may occur.

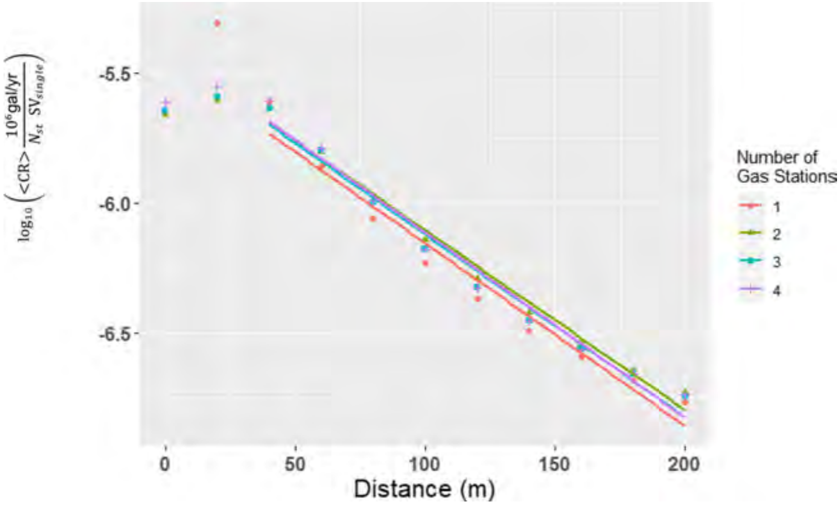

Fig. 6 Linear regression of the medians of lifetime cancer risk <CR> normalized by sales volume and then log-transformed for 1, 2, 3 and 4 gas stations. The regression excludes the first two distances (0 and 20 m)

This is because Eq. (2) represents a cancer risk averaged over all angles φ_i and because cancer risk may vary by more than an order of magnitude depending on φ_i for a given r (Fig. 5). Local meteorology and in particular variability in wind direction partially explain the spatial patterns and the directional dependence of modeled benzene concentrations, as a comparison between the wind rose (Supplemental Fig. 1) and the benzene concentrations fields (Figs. 3 and 4) shows. Therefore while Eq. (2) provides insights about how cancer risk depends on distance from gas station(s), detailed air dispersion simulations may be required to evaluate cancer risk for given receptor locations.

Equation for calculating cancer risk from gas station clusters

We proposed a simple equation, Eq. (2), which is based on an exponentially decaying function for estimating cancer risk as a function of distance from a gas station or a cluster of gas stations. Our statistical analysis (p-values and R^2) showed that cancer risk is a function of distance from gas station(s). Based on a theoretical premise, modeled cancer risk could be expected to scale linearly with sales volume SV_{single} but it was not clear whether it would also scale linearly with the number of gas stations N_{st}. One-way ANOVA, however, supports the hypothesis that cancer risk (averaged over all angles φ_i) scales linearly with total sales volume $\mathrm{SV}_{single}\ N_{st}$ for distances ≥40 m as evidenced by the similarity of the normalized cancer risk plots for the four different gas station configurations (Fig. 5) and the regression analyses for Eq. (2). However, Eq. (2) should not be used outside the range of distances r used to inform the regression (40 to 200 m).

As an example for an application of Eq. (2), we use it to calculate cancer risk at a distance $r = 150$ m from the aforementioned gas station dispensing 10 million gal/yr. With $a = -5.5$ and $b = -6.5\ \mathrm{km}^{-1}$, $\log_{10}(\ldots) = a + b\ r = -5.5 - 6.5 \times 0.15 = -6.5$, the same value determined from Fig. 6, thus also resulting in a cancer risk of 3 in a million.

Setback distances

Our Eq. (2), or variations thereof that account for actual emission rates and local meteorological conditions, provides a framework for formulating setback policies. E.g., if policy makers assume $\mathrm{CR} = 5 \times 10^{-6}$ is an acceptable cancer risk, one can solve Eq. (2) for r to calculate the distance at which this cancer risk is obtained, e.g., for a cluster of $N_{st} = 4$ gas stations having each a sales volume $\mathrm{SV}_{single} = 3.6$ million gal/year (or a single gas station dispensing 14.4 million gal/year): $r = \left[\log_{10}\left((\mathrm{CR})\frac{10^6\,\mathrm{gal/yr}}{N_{st}\ \ \mathrm{SV}_{single}}\right) - a)\right] / b = 145$ m. This distance can be interpreted as a setback distance, keeping in mind that cancer risk varies due to its directional dependence. This setback distance is much greater than the setback distance of 91 m recommended by CARB for California gas stations (with much lower emission factors) dispensing more than 3.6 million gal/year [26]. Thus, CARB guidelines should be used with caution if vapor emission control technology is below their standards.

Policy recommendations

While it is not surprising that cancer risks are higher for gas station clusters than for a single gas station, some policies on setback distances for gas stations account only for emissions from a single gas station [26], thereby neglecting the cumulative cancer risk arising from a cluster. We propose that policies should acknowledge the additional cancer risks arising from gas station clusters. This issue is of concern when a new gas station is built in an area where none is initially present and additional gas station(s) might be proposed thereafter or when a new gas station is built close to an existing one. Furthermore, our findings could provide a basis for improved standardization of policy at both the county and state level. Finally, we recommend that setback distances account for actual sales volume.

Limitations

Our study has some limitations. While we have devised an approach for estimating cancer risks from a gas station cluster, our study is not representative of any specific gas station development, because we only accounted for one set of meteorological conditions, assumed flat terrain, and made assumptions about fuel composition (benzene content) and emission prevention technology that are only representative of the US (except California). Indeed, according to an article published by the International Fuel Quality Center in 2009 benzene levels in gasoline can reach up to 7% in regions where these levels are regulated [39]; and levels can perhaps be even higher where not regulated. Moreover, benzene level may vary seasonally due to changes in fuel composition (winter versus summer fuel) [40–42] . However, because EPA [36] estimates of national gasoline benzene content (~ 1% by volume in 2016) and prior studies inform our assumptions, we feel they are a reasonable proxy. We also used emissions factors, which potentially underestimate actual emissions, as shown in a recent study that measured vent emissions at two fully functional US gas stations, finding that emissions greatly exceeded the emission factors listed in the CAPCOA (1997) study [24, 29].

Conclusions

We have developed a model to estimate cancer risk from gas station clusters, accounting for the increasing risk with additional gas stations and allowing for continuous rather than categorical sales volume inputs. Overall, we found that clusters of gas stations result in increased cancer risk compared to a single station. For instance, the cancer risk at 40 m for four gas stations each dispensing 1 million gal/year is 9.84×10^{-6} compared to 2.45×10^{-6} for one gas station. This framework can be utilized in real-life scenarios as a basis to estimate cancer risk by distance for gas station clusters in the US. Future work should consider developing a more general and widely applicable equation for cancer risk that also accounts for site-specific information such as emission factors, benzene content of the liquid gasoline and the gas phase in the ullage of the storage tank, and summary statistics of meteorological conditions. Future policies around setback distances should be reassessed to account for heightened risk from clusters.

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Data availability All data and material are publicly available.

Compliance with ethical standards

Conflicts of interest/Competing interests The authors declare they have no conflict of interest.

Code availability Code available upon request.

Ethics approval This study does not involve human subjects.

References

1. U.S. Energy Information Administration (EIA), "U.S. Product Supplied of Finished Motor Gasoline," Washington, DC: U.S. Energy Infromation Administration, 2019. [Online]. Available: https://www.eia.gov/dnav/pet/hist/LeafHandler.ashx?n=pet&s=mgfupus1&f=m.
2. Census. U.S., World Population Clock. United States Census Bureau, U.S. Department of Commerce. 2009.
3. Hilpert M, Breysse PN. Infiltration and evaporation of small hydrocarbon spills at gas stations. J Contam Hydrol. 2014;170:39–52. https://doi.org/10.1016/j.jconhyd.2014.08.004.
4. Hilpert M, Mora BA, Ni J, Rule AM, Nachman KE. Hydrocarbon Release During Fuel Storage and Transfer at Gas Stations: Environmental and Health Effects. Curr Environ Health Rep. 2015;2(4):412–22. https://doi.org/10.1007/s40572-015-0074-8.
5. Mora BA, Hilpert M. Differences in Infiltration and Evaporation of Diesel and Gasoline Droplets Spilled onto Concrete Pavement, Sustainability (Switzerland). 2017; 9: https://doi.org/10.3390/su9071271.
6. Morgester JJ, Fricker RL, Jordan GH. Comparison of Spill Frequencies and Amounts at Vapor Recovery and Conventional Service Stations in California. J Air Waste Manage Assoc. 1992;42(3):284–9. https://doi.org/10.1080/10473289.1992.10466991.
7. Agency for Toxic Substances and Disease Registry (ATSDR), "Interaction profile for benzene, toluene, ethylbenzene, and xylenes (BTEX)," Atlanta, GA: U.S. Department of Health and Human Services, Public Health Service, 2004.
8. Crossin R, Lawrence AJ, Andrews ZB, Churilov L, Duncan JR. Growth changes after inhalant abuse and toluene exposure: a systematic review and meta-analysis of human and animal studies," (in eng). Hum Exp Toxicol. 2019;38(2):157–72. https://doi.org/10.1177/0960327118792064.
9. Varjani SJ, Gnansounou E, Pandey A. Comprehensive review on toxicity of persistent organic pollutants from petroleum refinery waste and their degradation by microorganisms. Chemosphere. 2017;188:280–91. https://doi.org/10.1016/j.chemosphere.2017.09.005.
10. IARC. International Agency for Research on Cancer (IARC) monographs on the evaluation of carcinogenic risks to humans. vol. 100F, 2012. [Online]. Available: http://monographs.iarc.fr/ENG/Monographs/vol100F/.
11. Lan Q, et al. Hematotoxicity in workers exposed to low levels of benzene," (in eng). Science (New York, NY). 2004;306(5702):1774–6. https://doi.org/10.1126/science.1102443.
12. Morales Terrés IM, Miñarro MD, Ferradas EG, Caracena AB, Rico JB. Assessing the impact of petrol stations on their immediate surroundings. J Environ Manag. 2010;91(12):2754–62. https://doi.org/10.1016/j.jenvman.2010.08.009.
13. Jo WK, Oh JW. Exposure to methyl tertiary butyl ether and benzene in close proximity to service stations," (in eng). J Air Waste Manag Assoc (1995). 2001;51(8):1122–8. https://doi.org/10.1080/10473289.2001.10464339.
14. Jo W-K, Moon K-C. Housewives' exposure to volatile organic compounds relative to proximity to roadside service stations. Atmos Environ. 1999;33(18):2921–8. https://doi.org/10.1016/S1352-2310(99)00097-7.
15. Correa SM, Arbilla G, Marques MRC, Oliveira KMPG. The impact of BTEX emissions from gas stations into the atmosphere. Atmospher Pollut Res. 2012;3(2):163–9. https://doi.org/10.5094/APR.2012.016.
16. Hajizadeh Y, et al. Trends of BTEX in the central urban area of Iran: A preliminary study of photochemical ozone pollution and health risk assessment. Atmospher Pollut Res. 2018;9(2):220–9. https://doi.org/10.1016/j.apr.2017.09.005.
17. Hicklin W, Farrugia PS, Sinagra E. Investigations of VOCs in and around buildings close to service stations. Atmos Environ. 2018;172:93–101. https://doi.org/10.1016/j.atmosenv.2017.10.012.
18. Barros N, et al. Environmental and biological monitoring of benzene, toluene, ethylbenzene and xylene (BTEX) exposure in residents living near gas stations. J Toxic Environ Health A. 2019;82(9):550–63. https://doi.org/10.1080/15287394.2019.1634380.
19. Infante PF. Residential proximity to gasoline stations and risk of childhood leukemia. Am J Epidemiol. 2017;185(1):1–4. https://doi.org/10.1093/aje/kww130.
20. Steffen C, Auclerc MF, Auvrignon A, Baruchel A, Kebaili K, Lambilliotte A, et al. Acute childhood leukaemia and environmental exposure to potential sources of benzene and other hydrocarbons; a case-control study," (in eng). Occup Environ Med. 2004;61(9):773–8. https://doi.org/10.1136/oem.2003.010868.
21. Brosselin P, et al. Acute childhood leukaemia and residence next to petrol stations and automotive repair garages: the ESCALE study

(SFCE). Occup Environ Med. 2009;66(9):598–606. https://doi.org/10.1136/oem.2008.042432.

22. Harrison RM, Leung PL, Somervaille L, Smith R, Gilman E. Analysis of incidence of childhood cancer in the west midlands of the United Kingdom in relation to proximity to main roads and petrol stations," (in eng). Occup Environ Med. 1999;56(11):774–80. https://doi.org/10.1136/oem.56.11.774.
23. Baghani AN, et al. A case study of BTEX characteristics and health effects by major point sources of pollution during winter in Iran," (in eng). Environ Pollut (Barking, Essex : 1987). 2019;247:607–17. https://doi.org/10.1016/j.envpol.2019.01.070.
24. California Air Pollution Control Officers Association (CAPCOA), "Gasoline Service Station Industrywide Risk Assessment Guidelines," Toxics Committee of the California Air Pollution Control Officers Association (CAPCOA), 1997.
25. Wu B-Z, Hsieh L-L, Chiu K-H, Sree U, Lo J-G. Determination and impact of volatile organics emitted during rush hours in the ambient air around gasoline stations. J Air Waste Manage Assoc. 2006;56(9):1342–8. https://doi.org/10.1080/10473289.2006.10464589.
26. CalEPA. Air quality and land use handbook: a community health perspective. California Environmental Protection Agency & California Air Resources Board. 2005.
27. Mohai P, Kweon B-S. Michigan school siting guidelines: taking the environment into account. University of Michigan. 2020. [Online]. Available: https://deepblue.lib.umich.edu/handle/2027.42/156009.
28. Montgomery. Zoning text amendments: filling station - use standards ZTA-15-07. Montgomery County Council. 2015. [Online]. Available: https://www.montgomerycountymd.gov/COUNCIL/Resources/Files/zta/2015/20151201_18-07.pdf.
29. Hilpert M, Rule AM, Adria-Mora B, Tiberi T. Vent pipe emissions from storage tanks at gas stations: Implications for setback distances. Sci Total Environ. 2019;650:2239–50. https://doi.org/10.1016/j.scitotenv.2018.09.303.
30. NOAA/ESRL. NOAA/ESRL Radiosonde Database. [Online]. Available: https://ruc.noaa.gov/raobs/. Accessed 3 Nov 2018
31. EPA. User's Guide for the AMS/EPA Regulatory Model (AERMOD). vol. EPA-454/B-19-027, 2019.
32. EPA. User's Guide for the AERMOD Meteorological Preprocessor (AERMET). vol. EPA-454/B-19-028, 2019.
33. Cimorelli AJ, Perry SG, Venkatram A, Weil JC, Paine RJ, Wilson RB, Lee RF, Peters WD , Brode RW, Paumier JO. AERMOD: description of model formulation. U.S. Environmental Protection Agency, Office of Air Quality Planning and Standards, Emissions Monitoring and Analysis Division, vol. EPA-454/R-03-004, 2004. [Online]. Available: https://www3.epa.gov/scram001/7thconf/aermod/aermod_mfd.pdf.
34. Perry SG, et al. AERMOD: A Dispersion Model for Industrial Source Applications. Part II: Model Performance against 17 Field Study Databases. J Appl Meteorol. 2005;44(5):694–708. https://doi.org/10.1175/JAM2228.1.
35. Environmental Protection Agency (EPA), "Air Quality: Widespread Use for Onboard Refueling Vapor Recovery and Stage II Waiver. 2012; 77: 95.
36. EPA. Gasoline mobile source air toxics. U.S. Environmental Protection Agency. https://www.epa.gov/gasoline-standards/gasoline-mobile-source-air-toxics (accessed.).
37. EPA. Risk assessment guidance for superfund volume I: human health evaluation manual (Part F, Supplemental Guidance for Inhalation Risk Assessment. vol. EPA-540-R-070-002, 2009.
38. Seinfeld JH, Pandis SN. Atmospheric chemistry and physics: from air pollution to climate change. Hoboken: Wiley; 2016.
39. Poirier L, Vora K.. Top-100 Countries Ranked by. Standards for gasoline benzene limits al fuel.. Hart Energy Consulting, 2009. [Online]. Available: http://environmentportal.in/files/Gasoline%20benzene%20limits.pdf.
40. Gentner DR, Harley RA, Miller AM, Goldstein AH. Diurnal and Seasonal Variability of Gasoline-Related Volatile Organic Compound Emissions in Riverside, California. Environ Sci Technol. 2009;43(12):4247–52. https://doi.org/10.1021/es9006228.
41. Zimmerman N, et al. Field Measurements of Gasoline Direct Injection Emission Factors: Spatial and Seasonal Variability. Environ Sci Technol. 2016;50(4):2035–43. https://doi.org/10.1021/acs.est.5b04444.
42. Chin J-Y, Batterman SA. VOC composition of current motor vehicle fuels and vapors, and collinearity analyses for receptor modeling. Chemosphere. 2012;86(9):951–8. https://doi.org/10.1016/j.chemosphere.2011.11.017.