

Assessment of Occupational Exposure to VOCs at the Gantry Gasoline Terminal

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The gasoline components dispersed in the workplace environment have direct impact on human health because of their carcinogenic and mutagenic potential. The volatile portion of these compounds generally consists of a mixture of monoaromatics such as benzene, toluene, ethylbenzene, xylenes and substituted benzene compounds. Investigation of levels of these chemicals at the petroleum liquid storage, and distribution facilities is of growing importance for assessing the occupational exposure of workers at these locations. A continuous monitoring was conducted at the Gantry Terminal of a refinery with portable samplers for determining 8-hr TWA at various locations involving petroleum liquid loading and distribution in road tank trucks, rail cars, and drums. After thermal desorption, the concentrations were determined in the laboratory with advanced Varian Model 2200GC/MS system. TVOCs level ranged between 0.10 to 1.97 mg/m³. The 8-hr TWA for benzene, toluene, and xylenes were low and within the limits for occupational exposure of gasoline components.

Key words: VOCs, occupational exposure, 8hr-TWA, gasoline, BTEX, distribution, gantry

Introduction

Gasoline is a complex mixture of many components in variable proportions. Its composition varies from brand to brand and refinery to refinery. The composition of gasoline vapours is dominated by volatile organic compounds. Several groups of workers are exposed to this complex mixture repeatedly. Road tankers, rail cars and drum fillers are the heaviest workers exposed to these components. In addition, a vast number of other people are frequently exposed while refueling their vehicles.

A study was carried out in a Gantry Terminal of a Refinery located about 35 km from the Refinery. The terminal on an monthly average dispatches 11475 kL of motor spirit, 5153 kL of super kerosene oil, 21792 kL of high speed diesel, 2962 kL of LDO and 8845 kL of furnace oil. It has a facility for filling automatically 20 tankers simultaneously with the first 5 terminals for petrol (motor spirit), the next 6 terminals for diesel, next 5 for kerosene oil, and the remaining 4 for furnace oil. Each terminal requires about 30 minutes for loading the tankers. The terminal also has a separate drum filling facility for petrol, diesel and kerosene and a rail car loading facility for variety of fuels. Gasoline vapours escape into the atmosphere during bulk loading and storage of these products.

The workers at the terminals are exposed to a broad range of volatile organic compounds (VOCs). According to the amendments done in 1990 to the Clean Air Act in the United States, and European Commission Directive 63/94/EC (EC 1994), storage installations and loading and unloading equipment must be designed and operated in accordance with the technical provisions to reduce emissions of volatile organic

compounds. Measurement of these volatile organic compounds is of great importance in view of the health effects and occupational exposure at the workplace. The range of adverse effects due to these compounds include cancer and a number of other chronic disorders, e.g. aplastic anemia, pancytopenia, pulmonary (lung), structural changes, and a number of acute health disorders, e.g., dyspnea (difficulty in breathing), upper respiratory tract irritation with cough, conjunctivitis, neurotoxic effects, e.g., visual blurring, tremors, delirium, unconsciousness, coma and convulsions.

The paper reports the results of 8-hr shift (TWA) exposure measurement carried out at various locations in a Petroleum Liquid Storage and Distribution Terminal of a major refinery in India. Workers at these locations are daily exposed to vapors of several organic compounds such as benzene, toluene, xylenes and substituted benzenes. No occupational exposure data for workers engaged in the gasoline transport chain, such as inland terminals, service stations, exist for VOC concentrations in our country. In addition, a considerable proportion of the general population is frequently exposed to gasoline constituents during refueling at service stations. Hence, this study was carried out as a baseline study for assessing how much a worker would be exposed to VOCs during loading/unloading of gasoline products into road tankers, barrels and rail wagons.

A sampling methodology for the workplace environment has been developed using indigenous battery operated samplers. Simultaneous separation of the volatile mixture of compounds, and their identification and quantification were done by advanced GC/MS system.

Assessment of occupational exposure to VOCs at the gantry gasoline terminal

Methodology

Monitoring procedure

The sampling of organic vapors in the gasoline storage and distribution terminal was carried out using battery operated organic vapour sampler. Five such samplers were installed in the area. Due to high safety aspects on spot, according to continuous sampling during 8 - hours working period in the day time was carried out so as to obtain according to OSHA exposure limit an 8 hour Time Weighted Average (TWA) as per the following equation :

$$8 \text{ hr TWA} = \frac{(C_x T_x) + \dots (C_n T_n)}{8}$$

Where,

C_x = the concentration measured during time interval T_x ;

n = the total number of intervals measured , and

8 = the total time of exposure, in hours

Active breathing zone samples were collected on to activated carbon(AC) tubes.

A known volume of the air was passed through a specially designed activated charcoal tube for large loading of volatile compounds, at a flow rate of 100 mL/min. The flow rates of the pumps were measured before and after sampling. The sampling tube was placed close to the face of the person filling the tankers, rail cars and drums at respective filling terminals. During the shift, about 8-10 workers were investigated at each terminal. The volatile organic compounds were adsorbed on the activated charcoal at the sampling location and subsequently sealed for transporting to the laboratory for analysis . The sampling tubes were prepared as per ASTM: D 3686-95 Method¹

The sampling system consisted of pump, flow regulator, tubing and activated carbon tubes. The pumps were calibrated with a primary soap bubble method(Pandya, 1988)². The sampling system was assembled at the location where sampling was to be carried out. The AC tube was inserted into the sampling line, placing the backup section nearest to the pump. The pump was turned on and the flow was adjusted to the desired value of 100 mL/min. Sampling was carried out for a total interval of 8 hours at each location so as to obtain 8 hr TWA. This was done thrice. After each sampling period, the flow rate was rechecked, the pump turned off, and all the pertinent information was recorded on the field data sheet. The sample tubes were sealed with the caps and the tubes were labeled with the appropriate sampling site information to identify it. The sampling tubes were brought to the laboratory for thermal desorption and subsequent analysis using GC/MS Varian 2200 with Electron Impact Detector.

A reference standard mix 2 of volatile organic compounds was obtained from Supelco, USA. The mix contains 13 volatile compounds. Different concentrations of the reference standard mix were impregnated into three activated carbon tubes and each tube was separately thermally desorbed. 100mL of the sample from the thermal desorption unit was injected into the GC /MS and multipoint calibration curves for 13 compounds were prepared.

Instrumentation

A Varian 3800 GC coupled with Varian 2200 ion trap mass spectrometer (Varian, Walnut Creek, USA) was employed for the determination of analytes using electron ionization (EI) mode.

Tuning of the mass spectrometer is necessary to ensure consistent mass spectra for day-to-day work. In addition, the tuning process is used to obtain certain minimum response factors for key compounds that are used to check overall system performance.

The mass spectrometer was calibrated daily using the fragments of perfluorotributylamine at m/z 28, 69, 131,264, 414, 464, 502, and 614. Splitless injections of 100mL volume were carried out with a split/splitless programmable temperature injector(SPI), and a delay of 5 min. The ion trap, manifold and the transfer line were kept at 125^o, 40^o and 230^oC respectively.

Separations were performed on Varian Chrompack Capillary Column WCOT Fused Silica (30 m long, 0.25mm ID) CP-624 . Helium (ultra pure 99.99% with less than one ppm each of water, oxygen, and total hydrocarbons) was employed as a carrier gas. The flow was adjusted to 1.2 mL/min. The detector was programmed to scan over a mass range 40-450 amu.

Reagents and standards

The standards used in this study were procured from Supelco, USA. As the quality of chemical measurements depend critically on the use of reliable reference materials, these were used for calibration of instrumentation, checking performance of instrumentation against specification, qualitative identification of analyte or species being sought, and quantitative validation of measurement methodology. A quantitative relation over a range of observed responses was established correlating each of several known concentration to corresponding signal, thus yielding a response curve. Using the calibration curve, the value of the unknown analyte was determined. The base- peak ion was used for quantitation, and three qualifier ions were used for confirmation. After qualitative criteria were met, compound concentrations were calculated from multipoint calibration curves using external standard. Individual VOCs were identified through the combination of a retention time match with known standard and match of experimental EI mass spectrum, after background subtraction

to a reference spectrum of standard library spectrum (NIST Library, USA).

At least five sampling traps were spiked with a mixture of VOC standard to determine the method detection limit (MDL) of the system. The MDL ranged from 0.2 to 0.4 mg/m³ for the target VOCs. Mean Relative Standard Deviation (RSD) for all the compounds were less than 20 % (**Table 1**).

Table 1 : Method Detection Limit(MDL) and Relative Standard Deviation(RSD) for measured VOCs

Chemical	MDL mg/m ³	RSD
Benzene	0.3	13
Toluene	0.3	16
Ethylbenzene	0.2	12
m/p-Xylene	0.3	15
1,3,5,trimethylbenzene	0.3	13
1,2,4,trimethylbenzene	0.4	15
p-Isopropyltoluene	0.3	15
Butylbenzene	0.3	12
Naphthalene	0.2	10

Results and discussion

Gasoline vapors escape into the atmosphere during the bulk loading of the petroleum liquid mixture consisting primarily of aliphatic hydrocarbons in the range of 6, 7-11,12. Gasoline also contains some aromatic and other unsaturated hydrocarbons. The percentage compositions of sulfur, aromatics, olefins and benzene in petrol being handled in India and Europe are summarized in **Table 2**³. It is observed that stringent measures are being considered for benzene emissions in the country.

In order to assess the exposure of workers to VOCs in the Terminal area, two battery operated samplers were installed at the top of road tanker loading platform for filling diesel and kerosene respectively. Another sampler was installed at the drum filling facility for loading kerosene, petrol, diesel, etc. At the Gantry Terminal, a rail car yard was used for transportation of petroleum liquid. Sampling was also carried out in such an area where workers were engaged in loading gasoline in rail cars. In the sampling area, operators of vehicles as well as workers engaged in electrical, mechanical, gauging, tank loading and inspection are occupationally exposed to volatile organic compounds. A reference station was installed at the Gantry entrance security gate to monitor the background VOCs concentrations. All the sampling locations are shown in **Fig.2**. The meteorological conditions at the Gantry terminals were also recorded. The prevailing wind direction during the sampling was from north to east direction.

Table 2 : Specifications of petrol in India and Europe *

Parameters	Specifications				
	India 2000	Mukhop adhyay Committee 2005	Europe 2003	Europe 2003	USA RFG
Sulfur (% by weight)	0.1-0.5	0.03	0.015	0.005	0.002
Aromatics (% by volume)	No limit	45	42	35	25
Olefins (% by volume)	No limit	18	18	14-16	6
Benzene (% by volume)	3-5	1 (in 1 six metros)	1	1	0.8

*CPCB Newsletter "Parivesh", <http://www.cpcb.nic.in/feb02phy9.htm>

Most of the VOCs were released in atmosphere during filling of the tankers, barrels and the wagons. Identification and quantification of all the emitted volatile compounds were difficult due to limitations in preparing standards of these compounds. However, only those compounds (as per standard reference mix) were quantified whose calibration curve was prepared by GC/MS. The sum of mean concentrations of these at each sampling location was reported as Total Volatile Organic Compounds (TVOCs).

The exposed samples were brought to the laboratory and thermally desorbed, and analyzed with the gas chromatograph equipped with ion-trap detector(GC/MSD Varian 2200).

The identified compounds at one of the sampling sites located at the road tanker loading facility is shown in **Fig 3**. Major emissions in the terminal area were found from the compounds such as benzene, toluene, ethylbenzene and xylenes. Besides, some substituted benzenes and polyaromatic hydrocarbons were identified. The results of major components detected and quantified at various sampling locations in the Terminal area are summarized in **Table 3**. It is observed that during handling of gasoline products, the major concentrations of compounds emitted in the workplace are benzene and toluene. Benzene is considered to be the most toxic chemical in this group of VOCs and its carcinogenic effects have been widely accepted (Harrison, 1998⁴; Larsen, 1998⁵). The risk of leukemia after exposure to low concentrations of benzene has been debated over the past 20 years (Infante, 1997^{6,7}; Van Ralt, H.G.S, 1982⁸; Rinsky, 1987⁹; Brett, 1989¹⁰; Payman, 1990¹¹; Paustenbach, 1992¹²; Crump, 1994¹³; Utterback, 1995¹⁴).

Toluene is the most abundant volatile compound found near the road tanker-filling terminal. It is also recognised as a tracer of vehicular emissions(Heen *et al.*, 1999¹⁵) Benzene to

Assessment of occupational exposure to VOCs at the gantry gasoline terminal

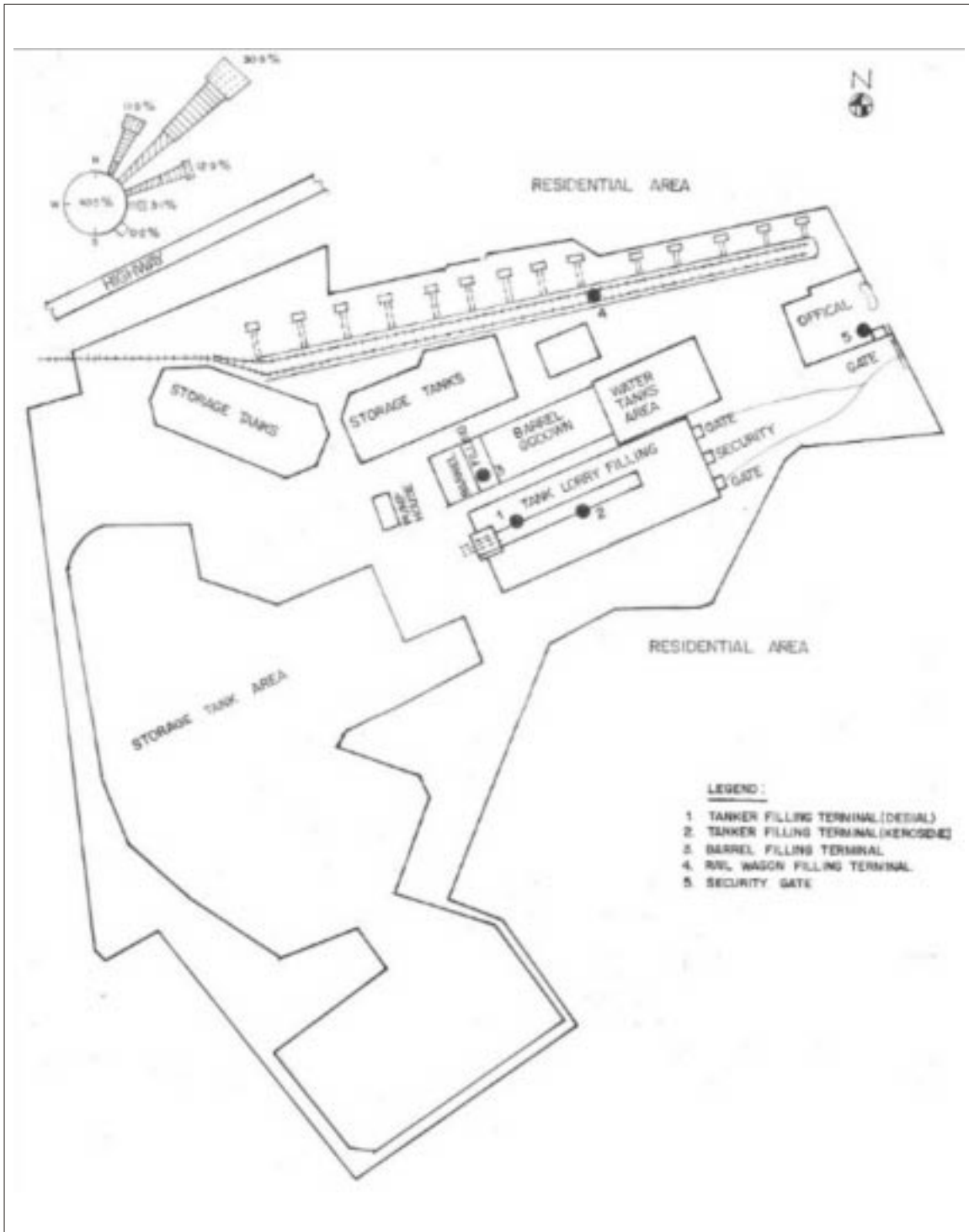


Fig 1: Sampling locations at the gasoline storage and distribution terminal area

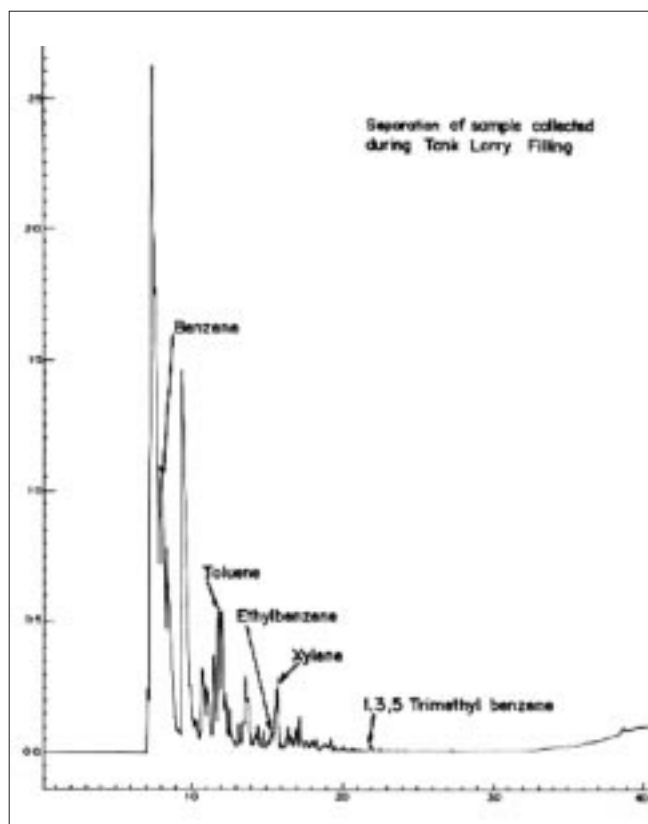


Fig 2 : Chromatogram of separated VOCs collected at road tank filling terminal

toluene ratio was determined for all the sampling sites in the terminal area. The ratio varies between 0.58 and 0.83. It has been observed that chemical removal of toluene in ambient air is much higher than benzene due to important reaction rates with OH radicals (Field, 1992¹⁶).

The emission of substituted benzenes, such as 135 trimethylbenzene and 124 trimethyl benzene, iso propyltoluene,

butyl benzene, has also been identified. These are mostly observed when the drums and rail wagons are being loaded with kerosene. 124 trimethyl benzene concentrations were comparatively higher among substituted benzenes. The concentrations of these compounds are summarised in **Table 3**. Polyaromatic compounds, like naphthalene, were also observed during loading operations of road tankers. Naphthalene is a part of light oil fractions and gets released into the atmosphere during handling of petroleum liquids.

Further analysis of the measured data enabled to compare the 8- hr TWA mean concentration ranges of benzene, toluene and xylenes (**Table 4**) with the occupational exposure limits of gasoline components of other countries (Concawe, 9/02). The occupational exposure limits (OELs) have been set nationally and internationally for many hazardous substances. These are referred to airborne concentrations of the hazardous substances. However, there is no widely accepted occupational exposure limit for gasoline vapour in Europe, but both The Netherlands and Sweden have established a national 8-hour TWA occupational exposure limit for gasoline vapour of 240 mg/m³ and 200 mg/m³ respectively. The occupational 8-hour TWA for workers exposed in the present study was low and within the limit.

The concentrations of Total Volatile Organic Compounds (TVOCs) were determined for each sampling location by adding the concentration of individual compounds measured at each sampling location. The results are summarized in **Table 5**. The TVOCs ranged between 0.109 to 1.971 mg/m³. The higher concentrations of TVOCs were observed near the road tanker loading areas. The lower TVOCs measured at the entrance security gate indicates that the occupational exposure is minimized as the distance from the loading area is increased. Further, occupational exposure varies and depends on the volatility of organic compounds.

Table 3 : Exposure data of VOCs (mg/m³)* as TWA- 8 hours for workers in various occupations transferring bulk gasoline

Component	Road tanker top loading at the terminal (Diesel)	Road tanker top loading (Kerosene)	Rail car top loading	Drum loading	Terminal security gate
Benzene	813.6 ± 28.2	303.1 ± 7.5	199.3 ± 10.0	192.2 ± 5.1	27.5 ± 3.5
Toluene	977.9 ± 7.5	516.5 ± 12.5	403.1 ± 7.9	261.8 ± 7.5	35.8 ± 7.0
Ethylbenzene	44.0 ± 4.0	53.3 ± 3.0	34.9 ± 2.0	40.6 ± 5.5	5.7 ± 3.0
m/p-Xylene	117.3 ± 7.0	162.9 ± 5.0	142.9 ± 7.0	152.3 ± 3.0	23.6 ± 2.0
1,3,5,trimethylbenzene	2.8 ± 2.0	8.4 ± 4.0	65.4 ± 5.0	108.4 ± 2.0	2.6 ± 1.5
1,2,4,trimethylbenzene	6.0 ± 2.0	12.4 ± 3.0	108.0 ± 7.0	272.3 ± 3.5	3.1 ± 1.5
p-Isopropyltoluene	BDL	2.7 ± 2.5	26.6 ± 5.5	64.8 ± 5.0	BDL
Butylbenzene	BDL	1.7 ± 2.6	15.9 ± 4.0	52.9 ± 4.0	BDL
Naphthalene	9.4 ± 3.5	28.1 ± 4.0	BDL	25.9 ± 6.0	10.7 ± 3.5

* Values are mean ± standard deviation (n=5)

Assessment of occupational exposure to VOCs at the gantry gasoline terminal

Table 4 : Comparison of measured 8-hrTWA with occupational exposure limits of gasoline components in mg/m³

Chemical CAS	Measured mean range 8-hr TWA in the present study	Finland	European Union	Sweden	United kingdom	The Netherlands	ACGIH (USA)
Gasoline	-	-	-	240	-	240	890
Benzene 71-43-2	0.19–0.81	9	3.25	1.5	16	3.2	1.6
Toluene 108-88-3	0.26–0.98	190	188	200	200	150	190
Xylenes 1330-20-7	0.11–0.15	440	221	200	441	210	440

Table 5 : Exposure data of TVOCs (mg/m³) as TWA-8hours for workers at road tanker, drum filling and rail car loading terminals

S.No.	Sampling location	Load at the terminal	Measured TVOCs, mg/m ³
1	Road tank filling	140 tanks daily	1.971
2	Road Tank filling		1.089
3	Drum filling	150 drums daily	0.996
4	Rail Car filling	Twice a week	1.171

A literature search about these compounds indicates that the concentrations of benzene in the atmosphere of cities, where chemical factories use or produce benzene, are more variable in USA. Benzene concentrations have been shown to vary between 0.4 and 16mg/m³ (Pellizzari,1982¹⁷). Levels of 3.2 mg/m³(1ppm) have been measured in the breathing zone during refueling of automobiles (Bond *et al.*,1986). In Frankfurt, Germany, the highest benzene levels have been measured in the vicinity of coke ovens(maximum 166.2 mg/m³, average 57.2 mg/m³), near industrial refineries (maximum, 102 mg/m³, average 13.4 mg/m³) and in congested traffic areas (maximum, 171.8 mg/m³, average, 16.9 mg/m³) (GDCh,1988¹⁸).

Besides benzene,a number of studies relating to exposure of xylenes have been reported. A number of studies have been performed on workers occupationally exposed to solvent mixtures including xylenes(Seppalainen ,1998¹⁹; Elofsan,1980²⁰; Lindstrom, 1982²¹; Maizlish, 1987²²; Vanvliet *et al*, 1987²³)

In a NIOSH study involving environmental monitoring of xylene in various worksites, the concentrations, where workers were exposed to gasoline and exhaust emissions ranged from < 0.08 to 68.3 mg/m³(0.02 to 15.7 ppm)(NIOSH, 1993a²⁴).

In USA, road tanker drivers were measured at 8 hour shift (Halder *et al*, 1986²⁵) during top and bottom loading with mean values of 46 to 90 mg/m³ for the total amount of hydrocarbons and 0.9 to 1.1mg/m³ for benzene.Short term benzene exposure in various occupations in Sweden were also reported (Nordlinger,1987²⁶).The mean exposure levels of road tanker drivers to benzene were 13 mg/m³ for loading and 1 mg/m³ for unloading (Berlin,1985²⁷). In Japan, the benzene exposure of road tanker drivers averaged to 0.38- 5.2 mg/m³ in short- term measurements and 0.04-0.07 mg/m³ in longer measurements, during loading with various techniques, and unloading (Kawait *et al.*,1996²⁸)

Conclusions

Loading losses are the primary source of evaporative emissions from rail car, road truck and drum filling operations. Gasoline vapors escape into the atmosphere during bulk loading of gasoline products and pose a health risk to workers, who inhale them during handling of the gasoline. The advancement in technology and introduction of vapor recovery systems have substantially reduced the exposure levels of the workers in the gasoline distribution chain. The measured 8 hour TWA values were found within the standard limits.

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Assessment of occupational exposure to VOCs at the gantry gasoline terminal

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