



INVESTIGATION OF VOLATILE ORGANIC COMPOUND (VOC) EMISSION IN OIL TERMINAL STORAGE TANK PARKS

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Abstract. The problem of VOC volatilization into the atmosphere from oil terminals is discussed in the paper. Investigation of VOC concentration in an oil terminal was performed in characterized spots of the main pollutant concentration – in storage tank parks of light and heavy oil products. A complex analysis of the impact of meteorological elements, oil-product-storage tank construction, the level of filling storage tanks with oil products on the emissions of VOCs in oil terminals and adjacent territories is provided in the paper. The dependence of VOC concentration on the following parameters of the construction of storage tanks was analysed: the capacity of storage tanks, the insulation between the wall of a storage tank and peculiarities of the pontoon construction (single, double, triple insulation). The results of the investigation may be applied for the development and improvement of the VOC calculation method (LAND 31-2007/M-11), reduction of VOC emissions in the existing oil terminals and when developing new ones.

Keywords: volatile organic compounds (VOCs), VOC emission, oil terminal, air pollution, vaporization.

1. Introduction

Today the environmental pollution research and modern environment protection technologies became a prior aspect not only in our country (Baltrėnas *et al.* 2008; Bimbaitė, Girgždienė 2007), but also all over the world.

According to the data records of the Department of Statistics to the Government of the Republic of Lithuania, the investment into environmental protection increased more than 12 times during the last year – from 15.1 (2001) to 187.7 million LTL (2006) (Paulauskienė *et al.* 2008).

The intensification and development of industrial processes has a negative impact on human health and environment. As a result, it increases waste product accumulation. It also has a disbalance of natural processes and reckless waste of natural resources. All of the above can cause greenhouse effect formation. Because of intensive expansion of energy, in industrial and transportation sectors, there is significant increase in atmosphere pollution in the last decade.

The western part of Lithuania is one of the most polluted areas by volatile organic compounds (VOCs), because it is a location of oil production and refinery industry as well as that of intensive development of oil terminals. The problem is becoming more serious and relevant when these objects are located near urban areas.

VOC emission from stationary pollution sources increased in Lithuania approximately 4 times – from 5.952 to 22.208 thousand t in the last years (1998–2006) (Paulauskienė *et al.* 2008).

Meanwhile, ambient air pollution by VOCs in Klaipėda increased approximately 6 times – from 0.564 to 3.551 thousand t (1998–2006). That's why major attention should be paid to this environmental problem of Klaipėda and its seaport. The presence of unpredictable meteorological conditions in the seaport area creates modern scientific research evaluation of ambient air pollution by VOCs (Laškova *et al.* 2007).

In oil terminals, VOCs enter the atmosphere from oil product storage and distribution systems (tanks, railway trestle, etc.), oil product reloading systems (Lashkova *et al.* 2007; Cetin and Odabasi 2003; Lin and Sree 2004).

When in the sunlight VOCs react with nitrogen oxides, the amount of ozone in the atmosphere increases and the formation of acid rain occurs (Aplinkos būklė 2004; Chiang *et al.* 2007; Murphy and Allen 2005). Hydrocarbons present in VOC composition, such as benzene, toluene, xylenes, are toxic, carcinogenic and harmful to human health (Pilidis and Karakitsios 2005; Ohura, Amagai 2006; Srivastava, Joseph 2006; Kerbach 2006, Hellen *et al.* 2005).

The evaporation of VOCs from oil and its products should be analysed not only from the environmental, but also from economic point of view. Through irreversible evaporation of oil products companies experience rather great quantitative and qualitative (the quality of oil and its products becomes worse) losses.

The aim of the present investigation is to carry out a test analysis of VOC concentrations in oil terminal storage tank parks through the evaluation of the type of a

product (light/heavy oil products), the level of an oil product in the storage tanks, the peculiarities of storage tank construction (capacity of storage tanks, type of insulation (single or double), the impact of the floatable pontoon) as well as the meteorological conditions.

2. Methodology of experimental investigation

The object of the present work is an oil terminal of AB “Klaipėdos nafta” that provides oil product transit services. This terminal is analogous to the terminals of other countries according to its technical and technological characteristics. Thus, the scheme offered for the research planning and the analysis of the results of the experiment may be used for carrying out research in other oil terminals.

Analyses of VOC concentration were made in Klaipėda University, Environmental Research Laboratory, using SHIMADZU GC-2010 gas chromatograph equipped with flame ionization detector (FID). At each observation post, air samples were put into Teflon bags. At observation posts, air samples were taken during as short time intervals as possible in order to avoid a significant fluctuation in meteorological elements.

A silicon capillary column, 0.5 m long with internal diameter of 0.52 mm was used for the quantitative analysis of VOC concentration. Temperature mode: evaporator – 125 °C; column – 125 °C; detector – 150 °C. Gas rate: helium – 30 ml/min, air – 400 ml/min, hydrogen – 40 ml/min.

Chromatograph is calibrated by n-hexane ($d^{20} = 0.6548 \text{ g/cm}^3$), while using a 20 l bottle. The bottle is hermetically sealed with a cork with a glass tube mounted

inside, through which n-hexane is let into the bottle or an air sample is taken. The calibration curve is made of 5 points; each of them is obtained having repeated the analysis 10 times.

The values of meteorological elements (air temperature, relative air humidity, atmospheric pressure, wind speed (in an altitude of 10 m) and wind direction) prevailing at the time when the air samples were taken from the Klaipėda meteorological station.

Disposition of observation points in the oil terminal territory is provided in Fig. 1. At one observation point, at least 3 air samples are taken, from which an average arithmetic value is obtained afterwards. Each result is a mean value of 9 measurements.

The following sampling sites were chosen for the measurement of VOC concentration within the territory of AB “Klaipėdos nafta” terminal: light (LOP) and heavy oil product (HOP) parks (on storage tanks next to breathers).

For the measurements of VOC concentration in the light oil (LOP) and heavy oil (HOP) product parks of the terminal of AB “Klaipėdos nafta”, the sampling was carried out by the breathers of the storage tanks.

Air samples were taken in eight supervision posts: 1 – LOP-10A, 2 – LOP-10B, 3 – LOP-5A, 4 – LOP-5B, 5 – HOP-32, 6 – HOP-20, 7 – HOP-5-3, 8 – HOP-5-5, 9 – HOP-5-4, 10 – HOP-5-6 (Fig. 1).

3. Results and discussion

Before choosing the zone for testing VOC concentration in the oil terminal, the distribution of the relevant pollutants in the whole territory of the tested object was analysed.

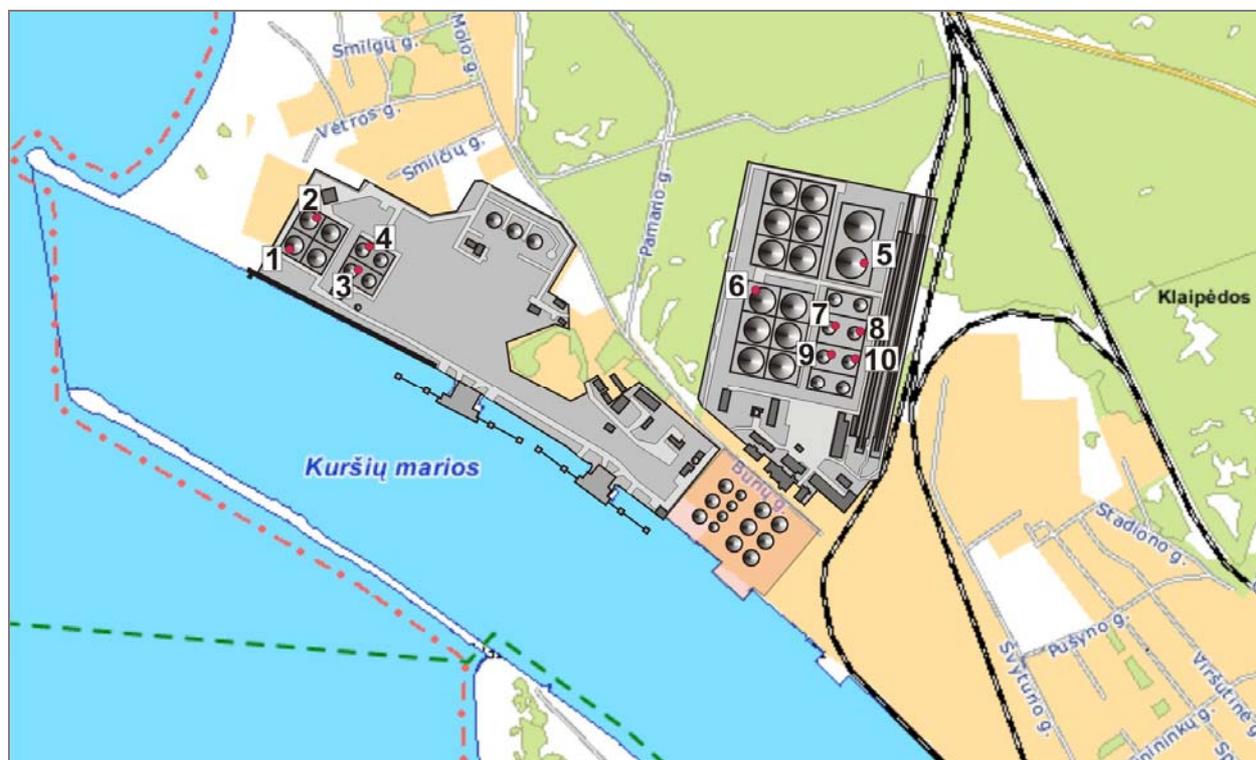


Fig. 1. Air sampling locations (at points from 1 to 10)

The results provided in Table 1 reveal that more than 60% of VOC are emitted in the zone of quays. More than one third of them are emitted into the environment from the storage tanks of light and heavy oil products, and only about 0.5% – from pump houses and overhead road. The smallest amount of VOCs (up to 0.1%) are emitted from the primary and water treatment equipment as well as other pollution sources; the total amount of pollutants here is less than 1 ton a year. In 2003, the total amount of VOC emissions from all the sources of the oil terminal was 4424.391 tons. This emission was dependent on various factors: storage-respiration/air, storage-evaporation, instantaneous emission, after opening the valves during loading works, due to poor sealing of the storage tanks, etc. (LAND 31-2007/M-11).

Light and heavy oil product parks, where about 38 % of the total amounts of VOCs are emitted, were chosen for the investigation.

Quite large amounts of VOCs are emitted into the ambient air from storage tank parks in the oil terminal. About 38% of oil product vapour are emitted into the ambient air when loading these products into the storage tanks and storing them. A detailed distribution of VOC emissions during the storage of different oil products in the storage parks of different types and capacity is provided in Table 2.

When determining the intensity of evaporation from the storage tanks, it is necessary to take into account the peculiarities of their construction, the oil level in the tanks, the type of oil and the prevailing meteorological conditions. The calculated results reveal that the largest amounts of VOCs are emitted in the quay and tank zones. These zones should be considered to be problematic ones, where more thorough investigation has to be carried out and possibilities to reduce air pollution have to be planned.

All the oil terminal storage tanks have stationary roofs; however, the internal floatable screen with circular sealants is installed not in all of them (Table 3). For example, such a preventive device is not installed in 5000 m³ and 20 000 m³ capacity HOP storage tanks. The floatable screens that inhibit the evaporation of the oil products are installed in all the other and newly-built HOP storage tanks of 32 000 m³ capacity.

It should be noted that not single, but double circular sealants are installed in all 10 000 m³ LOP and 32 000 m³ HOP storage tanks. The elements of such sealants enable to reduce VOC emissions in the storage tank parks.

VOC concentrations were tested on 16–26 October 2007. The meteorological conditions in the days of testing were also evaluated.

Fig. 2 shows the dominant wind speed and direction during air sampling. North-east wind with average speed of 2.7 m/s was dominating.

Table 1. VOC emissions in various technological zones in 2003 (calculations were carried out according to LAND 31-98/M-11 method)

Technological device/zone	Quantity, units	Work time, h/year	Emission from one unit, t/year	Total emission	
				t/h	%
Jetties	–	2236	2724.179	2.4367	61.5
Storage tanks	30	55395	1657.731	0.1097	37.5
Pump stations	3	26280	20.733	0.0008	0.5
Railway trestles	–	8760	17.882	0.0020	0.4
Other	–	91131	3.914	0.0001	0.1
Total:			4424.439	2.549	100

Table 2. VOC emissions from storage tanks in 2003 (calculations were carried out according to LAND 31-98/M-11 method)

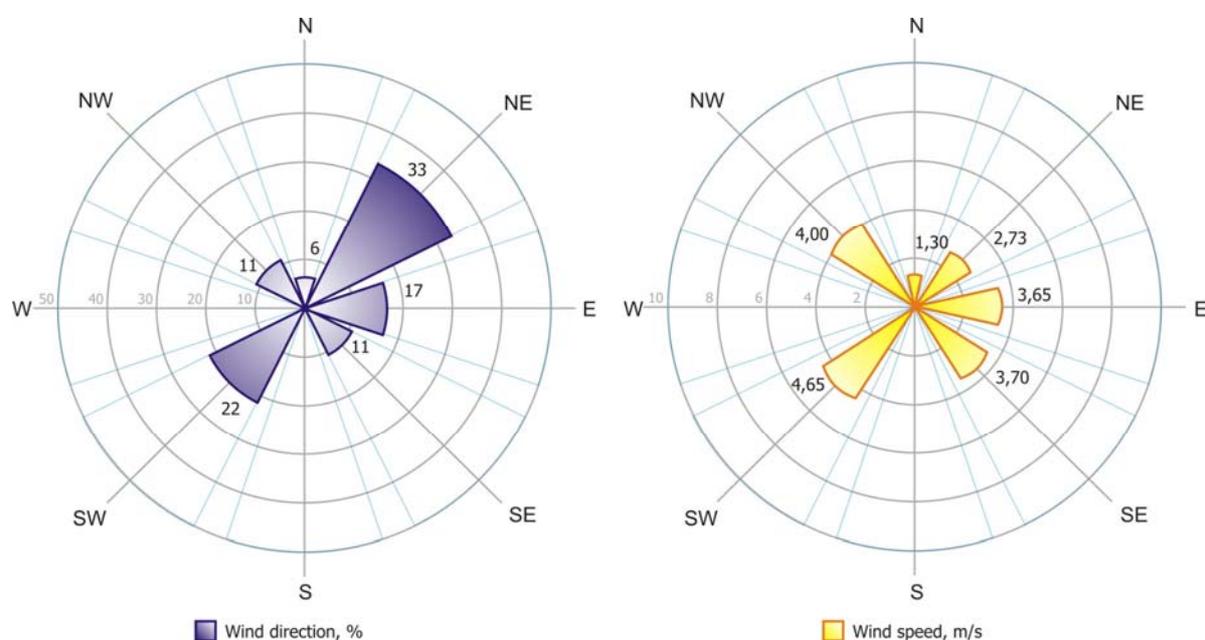
Technological device/zone	Quantity, units	Work time, h/year	Emission from one unit, t/year	Total emission	
				t/year	t/h
LOP storage tank of 10 000 m ³ (with pontoon and double insulation)	4	3641	43.042	172.18	0.0473
Fuel oil storage tank of 20 000 m ³ (without pontoon)	12	41422	–	1382.497*	0.0334
LOP storage tank of 5000 m ³ (with pontoon and single insulation)	4	3641	18.624	76.304	0.0210
Fuel oil storage tank of 5000 m ³ (without pontoon)	9	3366	2.675	24.075	0.0072
Boiler house diesel fuel storage tank of 5000 m ³ (without pontoon)	1	3325	2.675	2.675	0.0008
Total:				1657.731	0.109

* the amounts of VOCs increased, when the storage tanks were cleaned.

Table 3. Technical parameters of oil terminal storage tanks

Storage tank parks	Storage tank marking	Capacity, m ³	Height, m	Diameter, m	Height possible filling level, mm	Pressure/vacuum respiration valves, units	With stationary roof	With internal floatable screen	Seal topes	Length of circular sealant, mm
HOP	HOP-5-3	4799	11.7	22.9	11500	no	yes	–	no	–
	HOP-5-4	4773	11.7	22.9	11500			–	no	–
	HOP-5-5	4830	11.7	22.9	11500			–	no	–
	HOP-5-6	4813	11.7	22.8	11500			–	no	–
	HOP-20	19085	12.21	45	12192			yes	–	no
	HOP-32	33250	23.93	42	23293	no	yes	Double	131.88	
LOP	LOP-5A	5940	13.43	25	11690	3	yes	yes	Single	78.5
	LOP-5B	5940	13.43	25	11690				Single	78.5
	LOP-10A	10815	17.1	30	15300				Double	94.2
	LOP-10B	10815	17.1	30	15300				Double	94.2

Note: HOP – heavy oil products, LOP – light oil products

**Fig. 2.** Dominant wind speed and direction during air sampling

The air temperature alternated from 2.2 °C to 13.7 °C during the air sampling, and its mean value was about 7.2 °C (Fig. 3). Relative humidity ranges from 70% to 97% and atmosphere pressure ranges from 755 to 775 mm Hg.

It is determined that the VOC concentration value depends on the level of an oil product in the storage tank. The lower the level of an oil product in the storage tank, the bigger VOC concentration value is measured at the air valve. The alternation of the level of oil products in the storage tanks in the days of testing is provided in Table 4.

The following loading works were carried out in the days of testing:

No loading works were carried out during testing days 1–5 and 9, when air sampling was performed (Table 5).

During testing day 6, fuel oil was loaded at 273 t/h debit to the storage tank HOP-32 from the tank-wagons on railway tracks 1 and 2. Air sampling was performed at the beginning of the loading works. The final operation of the overcharging of the fuel oil to the tanker at low debit from the storage tank HOP-20 was carried out the same day.

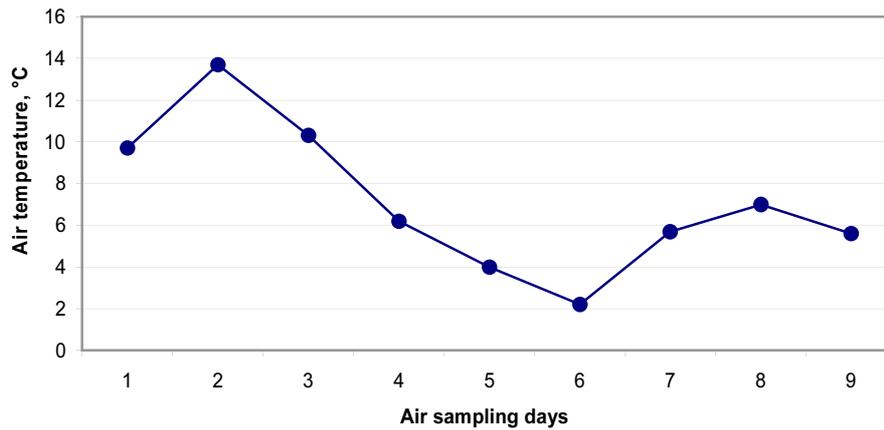


Fig. 3. Alternation of air temperature during air sampling

Table 4. Stored oil product level (m) in the storage tanks during air sampling

Air sam- pling days	Storage tank types/stored oil product level, m									
	LOP- 5A/ Jet A-1	LOP- 5B/ Jet A-1	LOP- 10A/ gasoline	LOP- 10B / gasoline	HOP-5- 3 / fuel oil	HOP-5- 4 / fuel oil	HOP-5- 5 / fuel oil	HOP-5- 6 / fuel oil	HOP- 20/ fuel oil	HOP- 32/ fuel oil
1			3.3						12.0	
2	3.6		1.6	4.2						
3	3.1	10.3	1.6	4.2						
4	2.1	10.3	1.1	4.2						
5							7.0		11.5	7.9
6							7.5		1.5	9.5
7										10.7
8					1.5	1.3	1.1	6.4	5.0	
9					1.5	1.3	1.1	6.4		

Table 5. Results of investigation on VOC concentration (mg/m³) in oil terminal storage tank parks

Air sampling days	Air sampling points									
	LOP- 5A	LOP- 5B	LOP- 10A	LOP- 10B	HOP-5-3	HOP-5-4	HOP-5-5	HOP-5-6	HOP- 20	HOP - 32
	VOC concentration (mg/m ³)									
1			5412.1* ↔						1084.5 ↔	
2	3.7 ↔		8.5 ↔	4.1 ↔						
3	2.8 ↔		41.9 ↔	3.6 ↔						
4	2.9 ↔	2.6 ↔	3.3 ↔	2.5 ↔						
5							493.1 ↔		1183.7 ↔	5.6 ↔
6							638.8 ↔		509.3 ↓	24.9 ↑
7										16.0 ↑
8					1469.6 ↔	48.4 ↓	2222.8 ↔	1870.4 ↔	624.3 ↑	74.4 ↑
9					1691.3 ↔		2155.9 ↔	461.7 ↔		

Note: Each result is a mean value of 9 measurements; * – pressure is 11 mbar, when the breather is open; ↑ – storage tank is loaded with an oil product, ↓ – an oil product is discharged from the storage tank, ↔ – an oil product is stored in the storage tank

During testing day 7, fuel oil was loaded (396 t/h) to the storage tank HOP-32 from the tank-wagons on railway tracks 1 and 2.

During testing day 8, fuel oil was loaded (314 t/h) to the storage tank HOP-32 from the tank-wagons on railway track 1. At the same time fuel oil was loaded (326 t/h) to the storage tank HOP-20 from the tank-wagons on railway track 2. Comparatively large concentrations of VOCs were recorded during the loading works to the latter storage tank, as there are no floatable screen and insulation devices installed in it. Fuel oil was also overcharged from the storage tank HOP-5-4 to the tanker the same day, thus a comparatively low VOC concentration was recorded at the latter storage tank.

We are going to analyse the results of alternation of VOC concentrations during testing.

The intensity of VOC evaporation from a storage tank depends directly on the construction parameters of the tank, the type of a loading operation (loading, unloading, storing), the type of a product (gasoline, Jet-A1, fuel oil, etc.), meteorological conditions and the level of a product in the storage tank. The volume of gases increases at the minimum filling level of the storage tank, thus the evaporation is then more intensive. And vice versa, the higher the filling level of oil product in the storage tank, the less VOC emissions.

This tendency may be observed, when analysing identical storage tanks LOP-5A and LOP-5B, where reactive Jet-A1-type fuel is stored (Tables 4, 5). Slightly higher VOC concentration is recorded in the first storage tank, the filling level of which is 3.5 times less in average (Tables 4, 5).

When analysing (testing days 2, 3 and 4) the dependence of VOC concentrations on the level of filling the identical storage tanks LOP-10 A and LOP-10 B (with identical characteristics of construction) with petrol, it can be seen that VOC concentration is 5 times bigger in the storage tank with the oil level lower more than three times (about 17.9 mg/m³).

When comparing the characteristics of construction of the storage tanks LOP-5A, B and LOP-10A, B, it should be noted that not only the capacity of the storage tanks differs, but also the parameters of their construction: a single sealing is installed in the storage tanks LOP-5A, B, while a double-sealing system is installed in the storage tanks LOP-10A and B, where more volatile products are stored.

The degree of filling with oil of the storage tank LOP-5A in the days of testing was averagely up to 25%, and the degree of filling of the storage tank LOP-10B was averagely 28% of the highest permissible filling level. Due to double sealing, VOC concentrations at the breathers of the storage tank LOP-10B were practically the same as those at the breathers of the storage tank LOP-5A (3.4 and 3.1 mg/m³, respectively; during testing days 2, 3 and 4). Thus, in certain cases, in order to reduce VOC emissions, it is recommended to store oil products in higher-capacity storage tanks with double or even triple sealing systems.

When the loading works were not performed, VOC concentration recorded at the breathers of the storage tanks HOP-5-3, 4, 5, 6 was alternating averagely between 1166 mg/m³ and 1581 mg/m³. The filling level of the storage tanks was alternating from 11% to 82%. Such big concentration values can be explained by the fact that there are no floatable pontoons and sealing systems installed in the storage tanks.

When the loading works were not performed, VOC concentration recorded at the breather of the storage tank HOP-20 was up to 1134 mg/m³ (testing days 1 and 5). At that moment, the filling level of the storage tank was 96%. The concentration specified is less than the one recorded at the storage tanks HOP-5-3, 4, 5, 6. VOC concentrations are less because there is an air valve installed in the roofing structure of the storage tank. In this case VOC emissions will be possible only when the pressure of VOC vapour exceeds the set limit value of the valve.

At the breathers of newly-constructed storage tanks (requirement of 94/63/EC directive) HOP-32 whose capacity came to 32 250 m³, as the fuel oil was being stored, an average of 38 mg/m³ was established. When testing the impact of the construction elements of the storage tanks on the VOC emissions from oil products into the ambient air, it was established that the most efficient measure for the reduction of VOC emissions in the oil terminals and the neighbouring territories is to install a floatable screen with a double sealing.

5. Conclusions

Summarizing the results of the performed testing, taking into account the type of a product, the level of an oil product in the storage tank, the characteristics of storage tank construction and meteorological conditions, the following conclusions can be drawn:

1. Having performed tests in the *light-oil product park*, it was determined that, when no loading works were carried out, VOC emissions at the storage tanks with a floatable screen were more hindered from the storage tanks, where double circular sealing systems were installed. This can be certified by the measurement results of VOC concentrations, that clearly show that average VOC concentrations (3.4 mg/m³) at the storage tank LOP-10B, that are two times bigger in size (10 000 m³) and have a double circular sealing system, is bigger only by about 8% than VOC concentration (3.1 mg/m³) at the 5 000 m³ storage tank LOP-5A with a single circular sealing system.

2. When loading works were carried out in the *heavy-oil product park*, VOC concentrations were alternating averagely between 1166 mg/m³ and 1581 mg/m³ at 5000 m³ storage tanks. This is due to the fact that no floatable screens and circular sealing systems are installed in these storage tanks, and the level of a product is low (up to 3.3 m). When 20 000 m³ storage tanks (HOP-20), where protective vacuum airing valves are installed, are filled with fuel oil, VOC concentration is up to 624.3 mg/m³. When no loading works were carried out, the lowest VOC concentration (38 mg/m³) was measured

at the breather of the storage tank HOP-32 that conforms to the requirements of the Standard 94/63/EC.

3. The results of testing and their complex analysis reveal how it is possible to reduce VOC emissions in oil-terminal storage-tank parks. The results of the present investigation may be used for the development and improvement of LAND 31-2007/M-11 method, when the value of VOC emission is assessed/measured not only according to the type of loading operation, but also according to the amount and type of sealing devices installed in the storage tanks.

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LAKIŲJŲ ORGANINIŲ JUNGINIŲ (LOJ) EMISIJOS TYRIMAS NAFTOS TERMINALO TALPYKLŲ PARKUOSE

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Santrauka

Analizuojama svarbi aplinkosauginė problema – oro tarša lakiaisiais organiniais junginiais (LOJ) naftos terminaluose. Šio darbo tyrimo objektas yra AB „Klaipėdos nafta“ terminalas, teikiantis naftos produktų tranzito paslaugas. Svarbu pažymėti, kad pagal technines ir technologines charakteristikas šis terminalas yra analogiškas kitiems terminalams pasaulyje. Taigi siūloma tyrimo planavimo bei eksperimento rezultatų analizės schema gali būti taikoma analogiškuose kitų terminalų tyrimuose.

Šio tyrimo tikslas – atlikti LOJ koncentracijos tyrimo naftos terminalo talpyklų parkuose analizę – įvertinti produkto rūši (šviesieji/tamsieji naftos produktai), talpyklose laikomo naftos produkto lygį, talpyklų konstrukcinius ypatumus (talpyklų tūrį, sandarinimo sistemas (viengubasis, dvigubasis, plūdrusis pontonas)) bei meteorologines sąlygas.

Tyrimo rezultatai parodė, kad LOJ emisiją galima sumažinti įrengiant talpyklose vidinius plūdruosius ekranus su žiedinėmis sandarinimo sistemomis. Kuo daugiau prevencinių priemonių, tuo labiau sumažinama LOJ emisija iš talpyklų. Pavyzdžiui, nevykdant krovos operacijos, LOJ koncentracija ties LOP-5 A, B talpyklų vakuuminiais vožtuvais vidutiniškai siekė 3 mg/m³. Šiose talpyklose įrengti plūdrieji ekranai ir viengubosios žiedinės sandarinimo sistemos. Nevyk-

dant krovos operacijos, LOJ koncentracija ties HOP-5 3–6 talpyklų alsuokliais siekė 1375 mg/m³. Šios talpyklos yra tik su nuostoviais stogais.

Iš kompleksinės gautų rezultatų analizės aiškėja, kaip pasiekti, kad LOJ emisija naftos terminalų talpyklų parkuose būtų mažesnė. Šio tyrimo rezultatai gali būti panaudoti LAND 31-2007/M-11 metodikai tobulinti, kai LOJ emisijos dydis vertinamas ne tik pagal atliekamos operacijos rūšį, bet ir pagal talpyklose įrengtų sandarinimo priemonių kiekį ir rūšį.

Reikšminiai žodžiai: lakieji organiniai junginiai (LOJ), LOJ emisija, naftos terminalas, atmosferos teršimas, garavimas.

ИССЛЕДОВАНИЕ ЭМИССИИ ЛЕГКОЛЕТУЧИХ ОРГАНИЧЕСКИХ СОЕДИНЕНИЙ (ЛОС) В ПАРКАХ РЕЗЕРВУАРОВ НЕФТЯНОГО ТЕРМИНАЛА

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Резюме

Обсуждается проблема загрязнения атмосферы легколетучими органическими соединениями (ЛОС) на нефтяных терминалах. Объектом исследования является АО «Клайпедос нафта», предоставляющее услуги по транзиту нефтепродуктов. Данное предприятие является типичным примером существующих нефтяных терминалов. Поэтому результаты исследования, а также организация эксперимента и анализ данных могут быть использованы для изучения проблемы испарения и распространения ЛОС на аналогичных нефтяных терминалах. Результаты исследования показали, что эмиссию ЛОС из парков резервуаров светлых и темных нефтепродуктов можно уменьшить за счет инсталлирования в резервуарах плавучих экранов с уплотнительными средствами. С увеличением количества превентивных мер уменьшаются выбросы ЛОС в атмосферу. Например, концентрация ЛОС (кроме операций погрузки) возле вакуумных клапанов LOP-5A в резервуарах светлых нефтепродуктов достигала в среднем 3 мг/м³. В эти резервуары были инсталлированы плавучие экраны и одинарные уплотнительные средства. Между тем концентрация ЛОС (кроме операций погрузки) возле дыхательного отверстия HOP-5-3 4-го, 5-го, 6-го резервуаров темных нефтепродуктов достигала в среднем 1375 мг/м³. В эти резервуары были инсталлированы лишь стационарные крыши. Результаты исследования и их анализ выявили способы уменьшения эмиссии ЛОС в зонах парков хранения светлых и темных нефтепродуктов. Результаты исследования могут быть также использованы для совершенствования методики LAND 31-2007/M-11, когда размер выбросов ЛОС будет рассчитываться с учетом не только типа операции, но также типа и количества уплотнительных средств, инсталлированных в резервуарах.

Ключевые слова: легколетучие органические соединения (ЛОС), эмиссия ЛОС, нефтяной терминал, загрязнение атмосферы, испарение.

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