Atmospheric release of organic solvents due to hazardous events in the paints and varnishes industry

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Abstract

Technogenic risk is the result of creation and development of hazards that originate from accidents in the technosphere. It involves emissions of hazardous industrial substances into the work and natural environments, fires, radioactive contamination, and contamination by toxic substances during their transport and storage. The qualitative and quantitative technogenic risk assessment has to be performed during the use, handling, transport, and storage of hazardous substances if it is decided that specific environmental elements at a specific location are likely to become exposed to hazardous substances, leading to environmental degradation. This paper discusses vulnerability zones – specifically, the high lethality zone and the irreversible effects zone – that are formed due to hazardous events or accidents in the paints and varnishes industry. Accidents are discussed in terms of exposure to solvents classified as hazardous to ecosystem components, the biosphere, anthroposphere, and the human population during their atmospheric dispersion. The vulnerability zones are designated using the REHRA (Rapid Environmental and Health Risk Assessment) methodology and their spatial arrangement is used to perform the risk assessment for the purpose of notifying authorized institutions and the public about a potentially increased risk at the hazard location and/or the immediate vicinity.

Keywords: reference distance; high lethality zone, irreversible effects zone, hazard.

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1. INTRODUCTION

Paints and varnishes industry, as a branch of the chemical industry, utilizes a wide variety of hazardous substances (toxic, flammable, and explosive) in production processes, which may cause chemical accidents. These accidents are accompanied with the risk of release of larger amounts of hazardous substances into the environment, leading to pollution and degradation of the affected environmental medium. Human health is most severely affected by outdoor ambient air pollution, which can be caused by fires, explosions, or propagation of toxic dispersing clouds generated during accidents by leaked substances used in the chemical industry.

The paints and varnishes industry utilizes different raw materials and intermediate goods, and technological procedures are so heterogeneous that production of paints and varnishes cannot be described in general terms. Operation of this industry creates different types of water, air, and soil pollution. Therefore, for any environmental risk assessment it is essential to know chemical composition of input as well as substances that leave the technological process in case and enter the environment.

Evaporation of a solvent and mixing of its vapour with air accelerates air flow and generation of dispersion clouds at longer or shorter distances from the emission point. Owing to their toxicity, solvents may have undesirable physiological effects on the exposed population if they exceed the maximum allowed concentrations for ambient air. There are considerable differences in toxic effects of solvents, ranging from fatal toxicity to delayed toxicity long after exposure [1]. Organic solvents are classified as hazardous substances, not only because of their toxicity but also because of their high flammability and explosiveness, which increase the risk of fires that propagate quickly and are not easily suppressed. In

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case of hazardous events, physicochemical properties of organic solvents entail different levels of risk both to the health of people exposed and to the environment.

Globally, there have been multiple hazardous events in recent years involving leaks and emissions of organic solvents with accompanying fires and/or explosions and deaths of dozens of people such as: acetic acid leak in Texas, USA, in 2021 [2]; vinyl chloride plant explosion in Mexico and ethylene glycol plant fire in Saudi Arabia, both in 2016 [3]; chemical industry accident of unknown origin in Taiwan in 2016, but subsequent analysis showed the presence of methanol, xylene, and butanol in the air [4]; alcohol explosion in a chemical plant tank in France in 2015, [5]; benzene leak in China in 2014, [6], and many more.

In Serbia, production of paints, varnishes, and other coatings, which involves organic solvents, takes place in about 150 plants with a continuous growing trend [7]. Serbian output was 128,119 t in 2018, 134,583 t in 2019, and 172,792 t in 2020 [8]. In recent years, following accidents in this branch of Serbian industry were recorded: Leskovac, 2006 [9]; near Niš, 2007 [10]; near Pirot, 2013 [11]; near Užice, 2019 [12]; and once again near Niš, 2020 [13].

This paper delineates vulnerability zones, specifically those with fatal outcomes and those with severe undesirable effects on humans and the environment, after paints and varnishes industry accidents involving leaks of organic solvents and formation of dispersion clouds. The vulnerability zone boundaries were calculated by using the REHRA (Rapid Environmental and Health Risk Assessment) methodology based on physicochemical properties and leaked quantities of organic solvents. This paper focuses solely on liquid organic solvents.

2. MATERIALS AND METHODS

2. 1. Organic solvents used in the paints and varnishes industry

Solvents used in the paint and varnish industry belong to different groups and classes of compounds, and they do not have common chemical properties. The simplest classification of solvents is into organic and inorganic solvents. Organic solvents are compounds that convert solid compounds or liquid non-volatile organic compounds into solutions without changing chemically. Organic solvents, which are used in the paint and varnish industry, are applied in mixtures, to which other diluents or fillers are added, to give the solution an appropriate viscosity. Organic solvents used in paints and varnishes are non-colloidal liquids that dissolve basic components and, when a film is formed, evaporate from the coating under normal conditions [14]. Regarding the physical properties, pure solvents are usually clear liquids, heavier or lighter than water, but their vapours are always heavier than air and closer to the ground. Solvent evaporation and mixing of the vapour with atmospheric air accelerates air flow. Physiological effects of organic solvents are produced only under specific conditions and at specific concentrations, which are taken as one of the factors of the solvent toxicity.

As the number and quantities of solvents increase, so does the level of hazard during the work. A large number and variety of solvents (different physical, chemical, and toxic properties) create particular problems in technical terms and in terms of occupational and environmental safety both under normal circumstances and during accidents. This leads to varying levels of health hazard to the exposed people in terms of type and severity of poisoning.

Organic solvents are also highly flammable and explosive substances, which is why their release or leak can cause quick-spreading fires that are difficult to suppress and that generate toxic dispersion clouds. During accidents involving the release or leaks of organic solvents, their volatility is the key property for determining environmental and health risk. Evaporation rate is an important physical property of solvents related to toxicity and it is usually, although not always, proportional to the boiling point. In addition to volatility, another important factor for assessing the hazard of organic solvent toxic effects is the boiling point and vapour pressure. The lower the boiling point of an organic solvent, the higher its volatility, which increases the probability of high vapour concentrations in the atmosphere.

Vapour pressure is also associated with substance toxicity. It is a well-known fact that, under normal external conditions, a highly toxic solvent with a low vapour pressure is less hazardous than a liquid with almost the same toxicity but with a lower vapour pressure. Some methodologies utilize the “risk index” (eq. 1) to assess the risk of organic solvent toxicity in the occupational and environmental atmospheres [15]:

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$$Ir = \frac{p}{\text{MDK}} \tag{1}$$

where *Ir* is risk index, *p* is vapoure pressure (101.325 kPa at 303 K) and MDK is maximum allowable concentration of organic solvent in the atmosphere, cm^3/m^3

It can be therefore concluded that only a complete study of physicochemical properties of organic solvents can indicate the hazards and harms during the work with these substances and determine the protective measures for occupational and environmental safety.

According to their level of hazard, organic solvents are classified as [16,17]:

- relatively benign solvents, which rarely harm human health under normal work conditions (petrol, ethyl chloride, ethyl acetate*, etc.*);
- low-toxicity solvents, which may affect health under specific work conditions (ethylene glycol, *etc.*);
- medium-toxicity solvents, which may cause permanent health harm under normal work conditions (toluene, xylene, amyl alcohol, trichloroethylene, *etc.*);
- high-toxicity solvents, which may cause permanent health damage in small amounts (benzene, carbon disulphide, methanol, nitrobenzene, *etc.*).

2. 2. Calculation of reference distance and potentially vulnerable zone upon release of organic solvents into the atmosphere

This paper relies on the REHRA methodology for rapid environment and health risk assessment pertaining to immediate effects of sudden release of organic solvents due to an accident in a paints and varnishes factory [16,17]. This methodology was developed for the primary purpose of enabling a quick assessment of the damage scope related to accidents that may originate in the chemical industry. The REHRA methodology is based on the use of a set of specific forms for the assessment and analysis of risks associated with accidents involving emission of hazardous substances. The results thus obtained are implemented in risk management procedures and in the selection of sites for industrial facilities during spatial planning and development of prediction techniques for emergency planning management in factories with increased likelihood of severe accidents. The methodology is also used to determine reference distance zones within which the unwanted effects of accidents are felt and to define vulnerability zone boundaries.

2. 2. 1. Reference distance calculation

Reference distance (*RD*) from the accident epicentre refers to boundaries of the zone within which fatalities among the exposed population are to be expected. The procedure for determining the RD involves integration of hazardous properties of substances (flammability, toxicity, explosiveness, etc.) and their released quantity due to an accident. This method relies on the use of a series of tables for assessment and management of technogenic and environmental risks. The procedure for determining the *RD* includes listing all hazardous substances in the hazard formulation stage of the risk assessment. Having been listed, the hazardous substances are then labelled and classified. The initial parameters for substance classification are the toxicity class and the volatility class. The toxicity class is based on the LC_{50} value of a substance (Table 1) [17]. LC_{50} is the median lethal concentration of a substance that in a defined time interval leads to the death of 50% of experimental animals.

Table 1. Toxicity classes (TOX) of substances based on $LC_{50\ \text{FOR RATS - (4H)}}$ [17]

Content, ppm	Toxicity class
0.01 – 0.1	8
0.1 – 1	7
1 – 10	6
10 – 100	5
100 – 1000	4
1000 – 10000	3
10000 – 100000	2

The volatility class is estimated based on the vapour pressure (p_V) at 20°C, boiling temperature (T_B) under normal conditions, or based on operating pressure (*p*), as shown in Table 2 [16,17].

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Table 2. Volatility classes (VL) of toxic substances [17]

Physical parameter	Volatility class
Toxic liquids	
$p_V \leq 5$ kPa	1
5 kPa$< p_V \leq 30$ kPa	2
$p_V > 30$ kPa	3
Toxic gases converted to liquid under pressure	
$T_B > -8.15$ °C	3
$T_B \leq -8.15$ °C	4
Toxic gases converted to liquid by freezing	
$T_B > -28.15$ °C	3
$T_B \leq -28.15$ °C	4
Toxic gases under pressure	
$p < 0.3$ MPa	2
0.3 MPa $\leq p < 2.5$ MPa	3
$p \geq 2.5$ MPa	4

The values shown in Tables 1 and 2 can be used to determine the toxicity class of substances in relation to LC_{50} and the volatility class. Table 3 shows the toxicity classes of substances according to the combination of LC_{50} and volatility class [16,17].

Table 3. Classification of toxic substances based on the sum of the toxicity (TOX) and volatility classes (VL) [17]

TOX + VL	CLASS
< 6	Low
7	Medium
8	High
9	Very high
10 - 12	Extreme

According to physicochemical characteristics of substances and their toxicity, the REHRA methodology provides groups of hazard categories for substances used in the chemical industry [18,19]. Table 4 provides an overview of organic solvents [18] used in the production of paints and varnishes [20], according to their chemical structure, and shows their hazard category according to the REHRA methodology [16,17].

Table 4. Groups of hazard categories of organic solvents used in the production of paints and varnishes [17]

Substance, CAS no.	Physicochemical properties	Group of hazardscat[a]
Aromatic hydrocarbons		
Benzene, 71-43-2	Flammable liquid with vapour pressure <30 kPa at 20°C (Flash point< 20°C)	1 & 3
Toluene, 108-88-3	Flammable liquid with vapour pressure <30 kPa at 20°C (Flash point< 20°C)	1 & 3
Xylene, 1330-20-7	Flammable liquid with vapour pressure <30 kPa at 20°C (Flash point> 20°C)	1 & 3
Naphthalene, 91-20-3	Flammable liquid with vapour pressure <30 kPa at 20°C (Flash point> 20°C)	1 & 3
Alcohols		
Methanol, 67-56-1	Flammable liquid with vapour pressure <30 kPa at 20°C (Flash point> 20°C)	1 & 3
Ethanol, 64-17-5	Flammable liquid with vapour pressure <30 kPa at 20°C (Flash point> 20°C)	1 & 3
Propanol, 71-23-8	Flammable liquid with vapour pressure <30 kPa at 20°C (Flash point> 20°C)	1 & 3
n-Butanol, 71-36-3	Flammable liquid with vapour pressure <30 kPa at 20°C (Flash point> 20°C)	1 & 3
Amyl alcohol, 123-51-3	Flammable liquid with vapour pressure <30 kPa at 20°C (Flash point> 20°C)	1 & 3
Cyclohexanol, 108-93-0	Flammable liquid with vapour pressure <30 kPa at 20°C (Flash point> 20°C)	1 & 3
Ethyleneglycol, 107-21-1	Flammable liquid with vapour pressure <30 kPa at 20°C (Flash point> 20°C)	1 & 3
Benzylalcohol, 100-51-76	Flammable liquid with vapour pressure <30 kPa at 20°C (Flash point> 20°C)	1 & 3
Ethers and acetals		
Ethyl ether, 60-29-7	Flammable liquid with vapour pressure >30 kPa at 20°C	4 & 6
n-Propyl ether, 108-20-3	Flammable liquid with vapour pressure <30 kPa at 20°C (Flash point> 20°C)	1 & 3
Ethyl ether, 60-29-7	Flammable liquid with vapour pressure <30 kPa at 20°C (Flash point> 20°C)	1 & 3

Substance, CAS no.	Physicochemical properties	Group of hazardscat[a]
Aldehydes and ketones		
Acetaldehyde, 75-07-0	Flammable liquid with vapour pressure > 30 kPa at 20°C	4 & 6
Acrolein, 107-02-8	Medium-toxicity liquids	18 & 21
Furfural, 98-01-1	Flammable liquid with vapour pressure <30 kPa at 20°C (Flash point> 20°C)	1 & 3
Acetone, 67-64-1	Flammable liquid with vapour pressure <30 kPa at 20°C (Flash point> 20°C)	1 & 3
Acetic acid, 64-19-7	Flammable liquid with vapour pressure <30 kPa at 20°C (Flash point> 20°C)	1 & 3
Formic acid, 64-18-6	Flammable liquid with vapour pressure <30 kPa at 20°C (Flash point> 20°C)	1 & 3
Esther		
Methylformate, 107-31-3	Flammable liquid with vapour pressure> 30 kPa at 20°C.	4 & 6
Ethylformate, 109-94-4	Flammable liquid with vapour pressure <30 kPa at 20°C (Flash point> 20°C)	1 & 3
Methyl acetate, 79-20-9	Flammable liquid with vapour pressure <30 kPa at 20°C (Flash point> 20°C)	1 & 3
Ethyl acetate,141-78-6	Flammable liquid with vapour pressure <30 kPa at 20°C (Flash point> 20°C)	1 & 3
Chlorinated hydrocarbons		
Chloroform, 67-66-3	Low-toxicity liquids (vapour pressure 21.3 kPa at 20°C)	16 & 17
Carbon tetrachloride, 56-23-5	Low-toxicity liquids (vapour pressure 11.9 kPa at 20°C)	16 & 17
Ethylchloride, 75-09-2	Flammable gas converted to a liquid under pressure	7 & 9
Dichloroethane, 107-06-2	Flammable liquid with vapour pressure <30 kPa at 20°C (Flash point> 20°C)	1 & 3
Trichloroethylene, 79-01-6	Low-toxicity liquids (vapour pressure 9.2 kPa bar at 20°C)	16 & 17
Tetrachloroethylene, 127-18-4	Low-toxicity liquids (vapour pressure 1.9 kPa bar at 20°C)	16 & 17
Chlorobenzene, 108-90-7	Flammable liquid with vapour pressure <30 kPa at 20°C (Flash point> 20°C)	1 & 3
Organic nitrogen compounds		
Aniline, 62-53-3	Flammable liquid with vapour pressure <30 kPa at 20°C (Flash point> 20°C)	1 & 3
Pyridine, 110-86-1	Flammable liquid with vapour pressure <30 kPa at 20°C (Flash point> 20°C)	1 & 3
Acetonitrile, 75-05-8	Flammable liquid with vapour pressure <30 kPa at 20°C (Flash point> 20°C)	1 & 3
Nitrobenzene, 98-95-3	Flammable liquid with vapour pressure <30 kPa at 20°C (Flash point> 20°C)	1 & 3
Organic sulfur compounds		
Carbondisulphide, 75-15-0	Flammable liquid with vapour pressure> 30 kPa at 20°C	4 & 6
Compounds with multiple functional groups		
Methylene glycol, 109-86-4	Flammable liquid with vapour pressure <30 kPa at 20°C (Flash point> 20°C)	1 & 3
Ethylene glycol, 107-21-1	Flammable liquid with vapour pressure <30 kPa at 20°C (Flash point> 20°C)	1 & 3
Diethylene glycol, 111-46-6	Flammable liquid with vapour pressure <30 kPa at 20°C (Flash point> 20°C)	1 & 3
o-Chloroaniline, 95-51-2	Flammable liquid with vapour pressure <30 kPa at 20°C (Flash point> 20°C)	1 & 3

[a] Groups of hazard categories are determined by the REHRA methodology [17].

According to the REHRA methodology, after a substance has been classified into a group of hazard categories, a reference hazard number is assigned depending on the method of storage. For instance, if a substance belongs to the hazard category group 1 & 3 and if it is stored in a storage facility inside a storage tank, its hazard reference number is 1, as shown in Table 5.

Table 5. Classification of types of hazardous substances and their storage methods [17].

Substance type	Substance characteristics	Ref. Hazard No.	Storage method
Flammable liquids	$p_V< 3\times10^4$ Pa at 20°C	[1]	Storage facility with a storage tank
		[3]	Other
Flammable liquids	$p_V> 3\times10^4$ Pa at 20°C	[4]	Storage facility with a storage tank
		[6]	Other
Flammable gases	Gas converted to a liquid under pressure	[7]	Storage above ground, truck or rail tank
		[9]	Other
Toxic liquids	Low-toxicity	[16]	Storage facility with a storage tank
		[17]	Other
Toxic liquids	Medium-toxicity	[18]	Storage facility with a storage tank
		[21]	Other

Reference numbers from Table 5 are combined with the maximum quantity that can realistically be released during a single accident according to the matrix shown in Table 6. The reference number points to a single row, while the maximum substance quantity that can be released points to a single column. The combination of the two factors in the matrix determines the category of the effect associated with the event under observation (specified by a letter and a Roman numeral). The propagation boundary is presented as a category marked with a capital letter of the alphabet, and the shape of the effect of the dispersion cloud propagation is marked with a Roman numeral (Table 7).

Table 6. Categories of vulnerability zones according to the distance from the hazard point of origin depending on the quantity of released hazardous substance (Q_i) *[16,17]*

REF.HAZ. NO.	Q_i / t								
	0-1	1-5	5-10	10-50	50-200	200-1000	1000-5000	5000-10000	>10000
[1]	-	-	-	-	-	AI	BI	BI	CI
[3]	-	-	-	AI	BI	CI	DII	X	X
[4]	-	-	-	-	-	BI	CII	CII	DII
[6]	-	-	-	BII	CII	DII	EII	X	X
[7]	-	AI	BI	CI	DI	EI	FI	X	X
[9]	-	BII	CIII	CIII	DIII	X	X	X	X
[16]	-	-	-	-	-	AII	AII	BII	CIII
[17]	-	-	-	AIII	AII	BII	CII	CII	CII
[18]	-	-	-	AIII	BIII	DIII	EIII	FIII	FIII
[21]	-	BII	CIII	DIII	EIII	FIII	FIII	X	X

Notes: X designates that this substance/quantity combination is not found in regular industry practice; – designates negligible effects

The combinations shown in Table 6 are elaborated in Table 7 in order to determine the category of damage (marked by a letter of the alphabet), reference distance (*RD*) that defines the boundaries of high lethality zones, estimation of the surface area potentially threatened by the accident (*DS*), and the shape of the effect from dispersion cloud propagation (*DF*), marked by the Roman numerals I, II, and III. In this way, the most precise value of the scope of vulnerability will be calculated by interpolating the data from Table 7 with the corresponding data referring to the ranges of released substance quantities (Table 6). This piece of data is particularly important for calculating the impact of hazards on all analyzed environmental components and the human population.

Roman numerals in Table 7 refer to the effect shape as a circle of the dispersion cloud propagation, which centre is the hazard point of origin; numeral II is a semi-circle whose centre is the hazard point of origin, facing leeward; and numeral III is 1/10 of a circle with a point of the hazard point of origin, also facing leeward.

Determination of the effect shape and vulnerability boundaries (Table 7) is essential for assessing the impact of the hazardous substances on all analyzed environmental elements and the human population so that preventive and safety measures can be defined for the designated zones of dispersion cloud propagation.

Table 7. Toxic substance propagation boundaries according to category, with reference distance

Category	*RD* / m	*DF*		
		I	II	III
		DS / ha		
A	0 - 25	0.2	0.1	0.02
B	25 - 50	0.8	0.4	0.1
C	50 - 100	3	1.5	0.5
D	100 - 200	12	6	1
E	200 - 500	80	40	8
F	500 - 1000	300	150	30
G	1000 - 3000	-	-	300
H	3000 - 10000	-	-	1000

2. 2. 2. Calculation of vulnerability zone boundaries after the release of an organic solvent

Assessment of risk severity for ecosystem components, the biosphere, the anthroposphere, and the human population due to a hazardous substance explosion is performed by means of determining two vulnerability zones.

The first vulnerability zone is the high lethality zone, within which fatalities among the exposed population and severe damage to ecosystem components and/or the biosphere and/or the anthroposphere are to be expected. The radius of this zone due to toxic substance dispersion is determined by the following equation [16,17]:

$$RD_\mathrm{d} = RD_\mathrm{min} + \frac{\left(Q - Q_{i_\mathrm{min}}\right)}{\left(Q_{i_\mathrm{max}} - Q_{i_\mathrm{min}}\right)}\left(RD_\mathrm{max} - RD_\mathrm{min}\right), \tag{2}$$

where: RD_d is a reference distance that is defined as the radius of the high lethality zone, which can be in the form of a full circle, semicircle or 1/10 of a circle (Tables 6 and 7) and is expressed in meters, RD_min – minimum reference distance, RD_max is maximum reference distance (Table 7), Q is released quantity (Table 7), $Q_\mathrm{i\,min}$ is minimum quantity value within the range of hazardous substance release for a given vulnerability zone category (Table 6) and $Q_\mathrm{i\,max}$ is maximum quantity value within the range of hazardous substance release for a given vulnerability zone category (Table 6).

Another zone that forms during accidents is the irreversible effects zone, within which the expected effects may be severe, but not sufficient to be lethal to the exposed human population. The scope of the irreversible effects zone is estimated by using the impact coefficient I (eq. 3). Multiplying the values of RD_d (high lethality zone radius) with impact coefficient I defines boundaries of the irreversible effects zone, formed due to accidents involving toxic non-flammable substances. The impact coefficient for flammable or explosive substances has the constant value of 2. However, for toxic non-flammable substances, the impact coefficient is a function of LC_{50}, according to the equation [16,17]:

$$I = 0.35 + 0.65\sqrt{\frac{LC_{50\,30\mathrm{min}}}{IDLH}} \tag{3}$$

where: $IDLH$ –'immediately dangerous to life or health' concentration of toxic non-flammable substance when inhaled [21,22].

3. RESULTS AND DISCUSSION

Table 8 shows calculated vulnerability zone radii for organic solvents used in the paints and varnishes production and classified in the hazard category group 1 & 3 (Table 4). Substances in this group are flammable liquids with vapour pressure lower than 30 kPa at 20 °C and a flash point lower or higher than 20 °C (Table 4). These substances are categorized as low-toxicity substances. Reference distance for these substances was calculated under the assumption that the substances are stored in a way that yields larger vulnerability zones in case of an accident than the zones when the substances are stored in storage tanks. The calculations were made for the least favourable storage conditions during an accident, corresponding to reference number 3 according to Table 5.

Table 9 shows calculated reference distances for leaks of flammable, toxic, and explosive organic solvents from the hazard category group 4 & 6 (Table 4), whose vapour pressure is higher than 30 kPa at 20 °C. Again, the reference distance is determined for the least favourable storage conditions for organic solvents used in production, corresponding to reference number 6 according to Table 5.

Table 10 shows calculated reference distances for leaks of flammable toxic gases that are converted into liquids under pressure for production purposes. These substances belong to the hazard category group 7 & 9 (Table 4). To calculate the reference distance, reference number 7 was used, referring to the storage of these substances above the ground or their transport in trucks or rail tanks.

Table 11 shows calculated vulnerability zone radii, *i.e*. reference distance, for low-toxicity organic solvents belonging to the hazard category group 16 & 17. The calculations are based on reference number 17 for the least favourable conditions of toxic dispersion cloud propagation after an accident.

Reference distance for medium-toxicity liquid organic solvents from the hazard category group 18 & 21 is calculated for the least favourable conditions of dispersion cloud propagation after an accident. The results are shown in Table 12.

Table 8. Reference distance (RD_d) at different released quantities of organic solvents (Q) belonging to the group of hazardous substances with the reference number 3 and their propagation surface area (DS)

Q from 10 to 500 t			*Q* from 510 to 1000 t			*Q* from 1100 to 5000 t		
Q / t	RD_d / m	*DS* / ha	*Q* / t	RD_d / m	*DS* / ha	*Q* / t	RD_d / m	*DS* / ha
10	0	0.2	510	69.375	3.0	1100	102.5	6.00
20	6.25		520	70		1200	105	
30	12.5		530	70.625		1300	107.5	
40	18.75		540	71.25		1400	110	
50	25		550	71.875		1500	112.5	
60	25	0.8	560	72.5		1600	115	
70	26.67		570	73.125		1700	117.5	
80	28.33		580	73.75		1800	120	
90	30		590	74.375		1900	122.5	
100	31.67		600	75		2000	125	
110	33.33		610	75.625		2100	127.5	
120	35		620	76.25		2200	130	
130	36.67		630	76.875		2300	132.5	
140	38.33		640	77.5		2400	135	
150	40		650	78.125		2500	137.5	
160	41.67		660	78.75		2600	140	
170	43.33		670	79.375		2700	142.5	
180	45		680	80		2800	145	
190	46.67		690	80.625		2900	147.5	
200	48.33		700	81.25		3000	150	
210	50.625	3.0	710	81.875		3100	152.5	
220	51.25		720	82.5		3200	155	
230	51.875		730	83.125		3300	157.5	
240	52.5		740	83.75		3300	157.5	
250	53.125		750	84.375		3400	160	
260	53.75		760	85		3500	162.5	
270	54.375		770	85.625		3600	165	
280	55		780	86.25		3700	167.5	
290	55.625		790	86.875		3800	170	
300	56.25		800	87.5		3900	172.5	
310	56.875		810	88.125		4000	175	
320	57.5		820	88.75		4100	177.5	
330	58.125		830	89.375		4200	180	
340	58.75		840	90		4300	182.5	
350	59.375		850	90.625		4400	185	
360	60		860	91.25		4500	187.5	
370	60.625		870	91.875		4600	190	
380	61.25		880	92.5		4700	192.5	
390	61.875		890	93.125		4800	195	
400	62.5		900	93.75		4900	197.5	
410	63.125		910	94.375		5000	200	
420	63.75		920	95				
430	64.375		930	95.625				
440	65		940	96.25				
450	65.625		950	96.875				
460	66.25		960	97.5				
470	66.875		970	98.125				
480	67.5		980	98.75				
490	68.125		990	99.375				
500	68.75		1000	100	6.00			

Table 9. Reference distance (RD_d) with the released quantity of organic solvents (Q) belonging to the group of hazardous substances with reference number 6 and their propagation surface area (DS)

Q from 10 to 500 t			*Q* from 510 to 1000 t			*Q* from 1100 to 5000 t		
Q / t	RD_d / m	*DS* / ha	*Q* / t	RD_d / m	*DS* / ha	*Q* / t	RD_d / m	*DS* / ha
10	25	0.4	510	138.75	6.00	1100	207.5	40.00
20	31.25		520	140		1200	215	
30	37.5		530	141.25		1300	222.5	
40	43.75		540	142.5		1400	230	
50	50		550	143.75		1500	237.5	
60	53.33	1.5	560	145		1600	245	
70	56.67		570	146.25		1700	252.5	
80	60		580	147.5		1800	260	
90	63.33		590	148.75		1900	267.5	
100	66.67		600	150		2000	275	
110	70		610	151.25		2100	282.5	
120	73.33		620	152.5		2200	290	
130	76.67		630	153.75		2300	297.5	
140	80		640	155		2400	305	
150	83.33		650	156.25		2500	312.5	
160	86.67		660	157.5		2600	320	
170	90		670	158.75		2700	327.5	
180	93.33		680	160		2800	335	
190	96.67		690	161.25		2900	342.5	
200	100		700	162.5		3000	350	
210	101.25	6.00	710	163.75	6.00	3100	357.5	
220	102.5		720	165		3200	365	
230	103.75		730	166.25		3300	372.5	
240	105		740	167.5		3400	380	
250	106.25		750	168.75		3500	387.5	
260	107.5		760	170		3600	395	
270	108.75		770	171.25		3700	402.5	
280	110		780	172.5		3800	410	
290	111.25		790	173.75		3900	417.5	
300	112.5		800	175		4000	425	
310	113.75		810	176.25		4100	432.5	
320	115		820	177.5		4200	440	
330	116.25		830	178.75		4300	447.5	
340	117.5		840	180		4400	455	
350	118.75		850	181.25		4500	462.5	
360	120		860	182.5		4600	470	
370	121.25		870	183.75		4700	477.5	
380	122.5		880	185		4800	485	
390	123.75		890	186.25		4900	492.5	
400	125		900	187.5		5000	500	
410	126.25		910	188.75				
420	127.5		920	190				
430	128.75		930	191.25	40.00			
440	130		940	192.5				
450	131.25		950	193.75				
460	132.5		960	195				
470	133.75		970	196.25				
480	135		980	197.5				
490	136.25		990	198.75				
500	137.5		1000	200				

Table 10. Reference distance (RD_d) with the released quantity of organic solvents (Q) belonging to the group of hazardous substances with reference number 7 and their propagation surface area (DS)

Q from 1 to 45 t			*Q* from 46 to 450 t			*Q* from 460 to 900 t			*Q* from 1000 to 5000 t		
Q / t	RD_d / m	*DS* / ha	*Q* / t	RD_d / m	*DS* / ha	*Q* / t	RD_d / m	*DS* / ha	*Q* / t	RD_d / m	*DS* / ha
1	0	0.2	46	95	0.3	460	297.5	80	1000	500	80
2	6.25		47	96.25		470	301.25		1100	512.5	300
3	12.5		48	97.5		480	305		1200	525	
4	18.75		49	98.75		490	308.75		1300	537.5	
5	25		50	100		500	312.5		1400	550	
6	30	0.8	60	106.67	12	510	316.25		1500	562.5	
7	35		70	113.33		520	320		1600	575	
8	40		80	120		530	323.75		1700	587.5	
9	45		90	126.67		540	327.5		1800	600	
10	50		100	133.33		550	331.25		1900	612.5	
11	51.25	3.0	110	140		560	335		2000	625	
12	52.5		120	146.67		570	338.75		2100	637.5	
13	53.75		130	153.33		580	342.5		2200	650	
14	55		140	160		590	346.25		2300	662.5	
15	56.25		150	166.67		600	350		2400	675	
16	57.5		160	173.33		610	353.75		2500	687.5	
17	58.75		170	180		620	357.5		2600	700	
18	60		180	186.67		630	361.25		2700	712.5	
19	61.25		190	193.33		640	365		2800	725	
20	62.5		200	200		650	368.75		2900	737.5	
21	63.75		210	203.75	80	660	372.5		3000	750	
22	65		220	207.5		670	376.25		3100	762.5	
23	66.25		230	211.25		680	380		3200	775	
24	67.5		240	215		690	383.75		3300	787.5	
25	68.75		250	218.75		700	387.5		3400	800	
26	70		260	222.5		710	391.25		3500	812.5	
27	71.25		270	226.25		720	395		3600	825	
28	72.5		280	230		730	398.75		3700	837.5	
29	73.75		290	233.75		740	402.5		3800	850	
30	75		300	237.5		750	406.25		3900	862.5	
31	76.25		310	241.25		760	410		4000	875	
32	77.5		320	245		770	413.75		4100	887.5	
33	78.75		330	248.75		780	417.5		4200	900	
34	80		340	252.5		790	421.25		4300	912.5	
35	81.25		350	256.25		800	425		4400	925	
36	82.5		360	260		810	428.75		4500	937.5	
37	83.75		370	263.75		820	432.5		4600	950	
38	85		380	267.5		830	436.25		4700	962.5	
39	86.25		390	271.25		840	440		4800	975	
40	87.5		400	275		850	443.75		4900	987.5	
41	88.75		410	278.75		860	447.5		5000	1000	
42	90		420	282.5		870	451.25				
43	91.25		430	286.25		880	455				
44	92.5		440	290		890	458.75				
45	93.75		450	293.75		900	462.5				

Comparison of propagation zone sizes or the calculated reference distances shown in Tables 8 to 12 is represented in Figure 1, which leads to a conclusion that under the same conditions of organic solvent release/leak, the largest high lethality zones are formed for the group of hazardous substances 18 & 21 with the reference number 21, while the smallest zones are formed for the group of hazardous substances 1 & 3, with the reference number 3, and the group 16 & 17, with the reference number 17.

Table 11. Reference distance (RD_d) with the released quantity of organic solvents (Q) belonging to the group of hazardous substances with reference number 17 and their propagation surface area (DS)

Q from 10 to 500 t			Q from 510 to 1000 t			Q from 1100 to 5000 t		
Q / t	RD_d / m	DS / ha	Q / t	RD_d / m	DS / ha	Q / t	RD_d / m	DS / ha
10	0	0.02	510	34.688	0.4	1100	51.25	1.5
20	1.3167		520	35		1200	52.5	
30	2.632		530	35.313		1300	53.75	
40	3.947		540	35.625		1400	55	
50	5.263		550	35.938		1500	56.25	
60	6.579	0.1	560	36.25		1600	57.5	
70	7.895		570	36.563		1700	58.75	
80	9.211		580	36.875		1800	60	
90	10.526		590	37.188		1900	61.25	
100	11.842		600	37.5		2000	62.5	
110	13.158		610	37.813		2100	63.75	
120	14.474		620	38.125		2200	65	
130	15.789		630	38.438		2300	66.25	
140	17.105		640	38.75		2400	67.5	
150	18.421		650	39.063		2500	68.75	
160	19.737		660	39.375		2600	70	
170	21.053		670	39.688		2700	71.25	
180	22.368		680	40		2800	72.5	
190	23.684		690	40.313		2900	73.75	
200	25		700	40.625		3000	75	
210	25.313	0.4	710	40.938		3100	76.25	
220	25.625		720	41.25		3200	77.5	
230	25.938		730	41.563		3300	78.75	
240	26.25		740	42.188		3400	80	
250	26.563		750	42.5		3500	81.25	
260	26.875		760	42.813		3600	82.5	
270	27.188		770	43.125		3700	83.75	
280	27.5		780	43.438		3800	85	
290	27.813		790	43.75		3900	86.25	
300	28.125		800	37.813		4000	87.5	
310	28.438		810	44.063		4100	88.75	
320	28.75		820	44.375		4200	90	
330	29.063		830	44.688		4300	91.25	
340	29.375		840	45		4400	92.5	
350	29.688		850	45.313		4500	93.75	
360	30		860	45.625		4600	95	
370	30.313		870	45.938		4700	96.25	
380	30.625		880	46.25		4800	97.5	
390	30.938		890	46.563		4900	98.75	
400	31.25		900	46.875		5000	100	
410	31.563		910	47.188				
420	31.875		920	47.5				
430	32.188		930	47.813				
440	32.5		940	48.125				
450	32.813		950	48.438				
460	33.125		960	48.75				
470	33.438		970	49.063				
480	33.75		980	49.375				
490	34.063		990	49.69				
500	34.375		1000	50	1.5			

Table 12. Reference distance (RD_d) with the released quantity of organic solvents (Q) belonging to the group of hazardous substances with reference number 21 and their propagation surface area (DS)

Q from 10 to 490 t			*Q* from 500 to 1000 t			*Q* from 1100 to 5000 t		
Q / t	RD_d / m	*DS* / ha	*Q* / t	RD_d / m	*DS* / ha	*Q* / t	RD_d / m	*DS* / ha
10	100	1.0	500	531.25	30.00	980	581.25	30.00
20	125		510	532.292		990	582.292	
30	150		520	533.33		1000	583.333	
40	175		530	534.375		1100	593.75	
50	200		540	535.417		1200	604.167	
60	220	8.0	550	536.458		1300	614.583	
70	240		560	537.5		1400	625	
80	260		570	538.542		1500	635.417	
90	280		580	539.5833		1600	645.833	
100	300		590	540.625		1700	656.25	
110	320		600	541.667		1800	666.667	
120	340		610	542.708		1900	677.083	
130	360		620	543.75		2000	687.5	
140	380		630	544.792		2100	697.917	
150	400		640	545.833		2200	708.333	
160	420		650	546.875		2300	718.75	
170	440		660	547.917		2400	729.167	
180	460		670	548.958		2500	739.583	
190	480		680	550		2600	750	
200	500		690	551.042		2700	760.417	
210	501.04	30.00	700	552.083	30.00	2800	770.833	
220	502.08		710	553.125		2900	781.25	
230	503.125		720	554.167		3000	791.667	30.00
240	504.168		730	555.208		3100	802.083	
250	505.208		740	556.25		3200	812.5	
260	506.25		750	557.292		3300	822.917	
270	507.2912		760	558.333		3400	833.333	
280	508.33		770	559.375		3500	843.75	
290	509.375		780	560.417		3600	854.167	
300	510.418		790	561.458		3700	864.5833	
310	511.458		800	562.5		3800	875	
320	512.5		810	563.542		3900	885.417	
330	513.542		820	564.583		4000	895.833	
340	514.583		830	565.625		4100	906.25	
350	515.625		840	566.667		4200	916.667	
360	516.667		850	567.7083		4300	927.083	
370	517.708		860	568.75		4400	937.5	
380	518.75		870	569.792		4500	947.917	
390	519.79		880	570.833		4600	958.333	
400	520.83		890	571.875		4700	968.75	
410	521.875		900	572.917		4800	979.167	
420	522.9167		910	573.958		4900	989.583	
430	523.958		920	575		5000	1000	
440	525		930	576.042				
450	526.042		940	577.083				
460	527.083		950	578.125				
470	528.125		960	579.167				
480	529.167		970	580.208				
490	530.208		980	581.25				

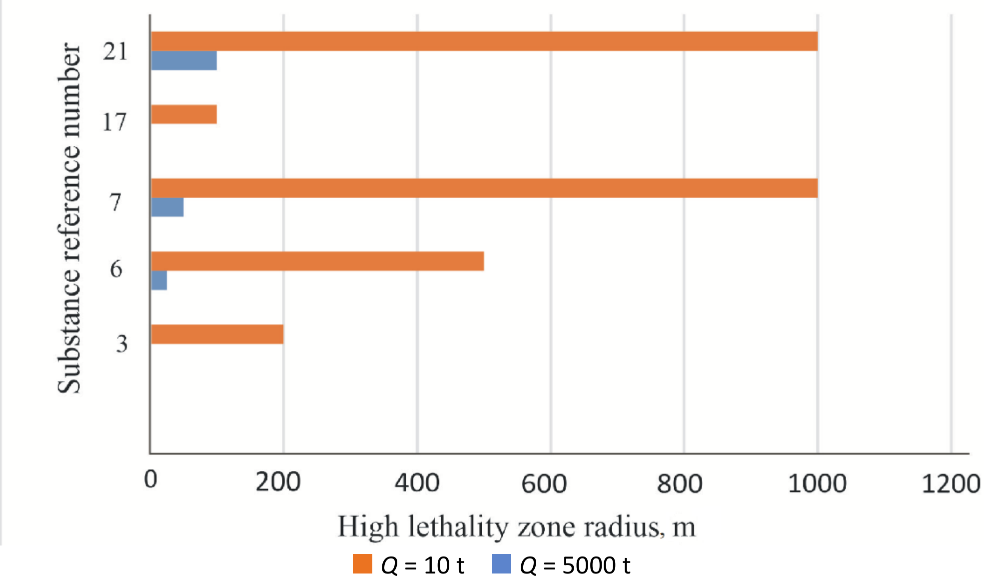

Figure 1. High lethality zone radii upon the release of Q = 10 t and Q = 5000 t of organic solvents according to their hazardous substance reference number

The impact coefficients (eq. 3) of toxic non-flammable substances that are used as organic solvents in paints and varnishes production are given in Table 13.

Table 13. Impact coefficient, I, and reported LC_{50} and IDLH values [22] of toxic organic solvents in the paints and varnishes industry

Non-flammable organic solvent	Impact coefficient	$LC_{50\text{FOR RATS - (0,5h)}}$ / ppm [22]	*IDLH* /ppm [22]
Acrolein	3.68	131 – 0.5 h	5
Chloroform	4.38	19.23 – 0.5h	500
Carbon tetrachloride	6.31	16.80– 0.5 h	200
Trichloroethylene	5.49	62.59– 0.5h	1000
Tetrachloroethylene	5.99	11.32– 0.5 h	150

After determining the reference distance (RD_d) of dispersion cloud atmospheric propagation from the accident point of origin and determining the impact coefficient (*I*), the following step is to map vulnerability zones and delineate the high lethality zone and the irreversible effects zone. Figure 2 shows an example of a map of the high lethality zone (orange circle on the map), which was calculated and shown in Table 8 above for the hypothetical leak of 120 t of benzene in a model paints and varnishes factory. The yellow doughnut in Figure 2 delineates the irreversible effects zone for this hypothetical event. The irreversible effects zone boundaries were obtained by means of multiplying the reference distance (RD_d) by 2, since benzene is a flammable substance (Table 4). Benzene dispersion has a circular shape for the 120 t leak since it is a toxic substance in the hazard category group 1 & 3 (Table 4) with the reference number 3 (Table 5), so that the shape of propagation (*DF*) is marked by the Roman numeral I according to Table 7.

Figure 3 shows another example of a map for the hypothetical leak of 250 t of carbon tetrachloride in the same model paints and varnishes factory. The shape of dispersion propagation of this substance, classified in the hazard category group 16 & 17 (Table 4) with the reference number 17 (Table 5), is a semi-circle with a centre at the point of origin of the leak and facing leeward, which is marked by the Roman numeral II according to Table 7. The reference distance for the leak of organic solvents in group 16 & 17 with the reference number 17 is given in Table 11, while the calculated impact coefficient for carbon tetrachloride is given in Table 13. The irreversible effects zone boundaries for this leak were obtained by multiplying the reference distance by the impact coefficient provided in Table 13.

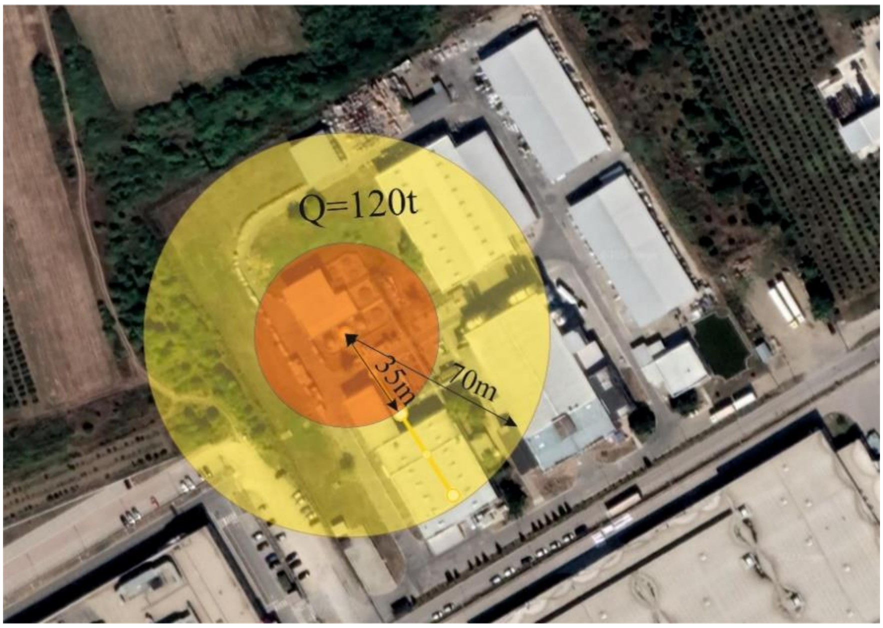

Figure 2. High lethality zone (orange) and irreversible effects zone (yellow) for a hypothetical leak of 120 t of benzene in a model paints and varnishes factory

Figure 3. High lethality zone (orange) and irreversible effects zone (yellow) during the hypothetical leak of 250 t of carbon tetrachloride in a model paints and varnishes factory

The shape of hazardous dispersion propagation of a substance emitted into the atmosphere during an accident may also be 1/10 of a circle, in addition to a full and semi-circle, with the release/leak point of origin facing leeward. Figure 4 shows the map of a hypothetical leak of 5,000 t of acrolein (C_3H_4O) in a model paints and varnishes factory. Acrolein belongs in the 18 & 21 group of hazardous substances (Table 4). For the leak of 5,000 t under conditions defined by the reference number 21, the reference distance is given in Table 12 and the impact coefficient in Table 13. As in the previous case, the irreversible effects zone was determined by multiplying the reference distance by the impact coefficient, and it is shown in yellow in Figure 4, while the high lethality zone is shown in orange. The shape of hazardous propagation is marked by the Roman numeral III (Table 7) and defined as 1/10 of a circle with the release and/or leak point of origin facing leeward.

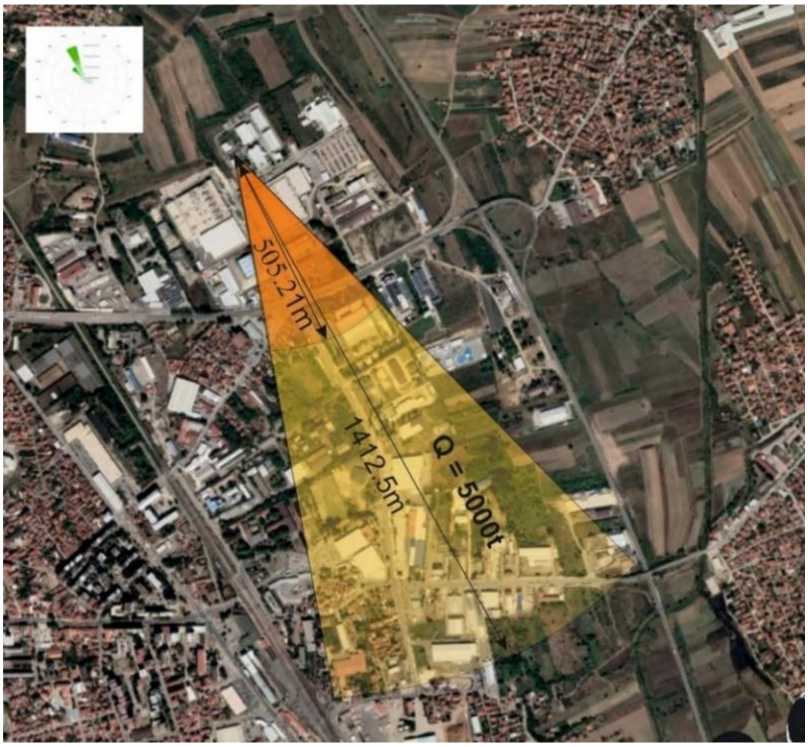

Figure 4. High lethality zone (orange) and irreversible effects zone (yellow) during the hypothetical leak of 5000 t of acrolein in a model paints and varnishes factory

Every possible case of release or leak of organic solvents used in paints and varnishes production can be represented on a map based on their calculated reference distances, given in Tables 8-12, and their calculated impact coefficients, given in Table 13. The map representation has practical implications for design analysis and performing the environmental impact assessment study, as it can help predict vulnerability zone propagation through urban areas. Another practical benefit of mapping vulnerability zones pertains to accident events, when it is necessary to determine the vulnerability zone territory in which appropriate safety and mitigation measures should be implemented.

4. CONCLUSION

Processes in chemical industry are inherently not absolutely safe and potentially pose a threat of chemical hazards. During hazardous events in this industry, there is increased likelihood of larger amounts of hazardous substances being released and causing fires, explosions, or toxic gas dispersions. There have been multiple occasions worldwide where chemical accidents happened due to the leak and emission of organic solvents that in turn resulted in fires and/or explosions, killing dozens of people. Since there is a justified need, both globally and in Serbia, for the production of paints, varnishes, and other coatings that require organic solvents, it is necessary to predict dimensions of toxic dispersion clouds propagating after the potential release/leak of these solvents during hazardous events. Severity of the effects of organic solvent explosions on ecosystem components, the biosphere, anthroposphere, and human population depends on physicochemical properties and toxicity of these solvents, as well as on the quantities that form the dispersion clouds. After the analysis of vulnerability zones formed during hazardous events in paints and varnishes production, using the REHRA methodology, the following conclusions can be drawn.

- Atmospheric release/leak of organic solvents, which are flammable liquids with vapour pressure lower than 30 kPa at 20 °C and flash point below or over 20 °C, and their dispersions will form high lethality zones that will be smaller than those formed due to release of other solvents under the same conditions. The shape of dispersion propagation is a circle or a semi-circle with the point of origin as the centre and facing leeward. The high lethality zone radius ranges from 0 to 200 m during the release/leak of 10 to 5,000 t of such a solvent. There are considerably more organic solvents with these properties in use in paints and varnishes production than any other organic solvents.

- Comparison of calculated sizes of high lethality zones due to organic solvent release/leak showed that the largest zones would be created due to hypothetical acrolein release/leak. The dispersion cloud is in the shape of 1/10 of a circle facing leeward from the point of release/leak. When the solvent is released/leaked in the quantity from 10 to 5,000 t, the high lethality zone radius ranges from 100 to 1,000 m.
- When emitted, chloroform ($CHCl_3$), carbon tetrachloride (CCl_4), trichloroethylene (C_2HCl_3), and tetrachloroethylene (C_2Cl_4) form a dispersion cloud that propagates in the shape of 1/10 of a circle facing leeward from the point of release/leak. The high lethality zone radius ranges from 0 to 100 m when these solvents are released/leaked in the quantity from 10 to 5,000 t.
- When emitted, ethyl ether ($C_4H_{10}O$), acetaldehyde (C_2H_4O), methyl formate ($C_2H_4O_2$), and carbon disulphide (CS_2) form a dispersion cloud that propagates in the shape of a semi-circle facing leeward. The high lethality zone radius ranges from 25 to 500 m when these solvents are released/leaked in the quantity from 10 to 5,000 t.
- When emitted, ethyl chloride (C_2H_5Cl), which is stored above ground or transported in truck or rail tanks forms a circular dispersion cloud in the atmosphere. The high lethality zone radius ranges from 0 to 1,000 m when ethyl chloride is released/leaked in the quantity from 10 to 5,000 t.

Knowledge of dispersion propagations of the analyzed hazardous organic solvents used in the paints and varnishes industry, as well as in other industries, and their hazard mapping have practical implications in accident prevention and implementation of safety and mitigation measures. Additionally, mapping of dispersion propagation with the designation of vulnerability zones has also practical implications for design analysis and environmental impact assessment for project proposals in process plant design, because it is used to predict hazardous substance dispersion propagations through urban areas.

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Disperzija organskih rastvarača u atmosferi pri hazardnom događaju u industriji boja i lakova

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Izvod

Tehnogeni rizik proizilazi iz opasnosti koje nastaju pri akcidentima u tehnosferi i obuhvata emisije opasnih materija iz industrije u radnoj i životnoj sredini, požare, kontaminaciju radioaktivnim supstancama, kao i kontaminaciju toksičnim supstancama pri njihovom transportu i skladištenju. Kvalitativno-kvantitativnu procenu tehnogenog rizika potrebno je sprovoditi pri upotrebi, rukovanju, transportu i skladištenju opasnih supstanci, ukoliko se zaključi da postoji mogućnost da će na određenoj lokaciji doći do izlaganja okoline tim supstancama što će dovesti do degradacije životne sredine. U radu je dat prikaz zone ugroženosti koja se formira u vidu zone visoke smrtnosti i zone ireverzibilnih efekata usled hazardnih ili akcidentnih događaja u industriji boja i lakova. Akcidentni događaji se posmatraju sa aspekta formiranja ekspozicije rastvarača pri njihovoj disperziji u atmosferi, a koji se svrstavaju u grupu opasnih supstanci po komponente ekosistema, biosferu, antroposferu i ljudsku populaciju. Zona ugroženosti se u radu utvrđuje primenom metodologije REHRA, a njihovo prostorno definisanje omogućava procenu rizika sa ciljem obaveštavanja nadležnih institucija i javnosti o mogućem povećanom riziku na mestu hazarda i/ili neposrednoj okolini.

Ključne reči: referentna udaljenost, zona visoke smrtnosti, ireverzibilna zona, hazard

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