The impact of municipal solid waste landfills in Suceava County on air quality

Dumitru MIHĂILĂ^{1*}, Valeria DIȚOIU¹ and Petruț-Ionel BISTRICEAN²

¹ "Stefan cel Mare" University of Suceava, Department of Geography

² National Meteorology Administration, CMR Moldova - Iasi, Suceava Weather Station, Romania

* Correspondence to: Dumitru Mihăilă, "Ștefan cel Mare" University of Suceava, Department of Geography. E-mail: mihaila_dum@yahoo.com.

©2014 University of Suceava and GEOREVIEW. All rights reserved. doi: 10.4316/GEOREVIEW.2014.24.1.125



Article history Received: February 2014 Received in revised form: June 2014 Accepted: July 2014 Available online: August 2014 **ABSTRACT:** The location of municipal solid waste (MSW) landfills in inappropriate places is a serious risk to the quality of all environmental factors. These waste disposal sites can become major sources of air quality deterioration through emissions of toxic gas resulted from anaerobic decomposition of organic waste. The paper discusses in detail the qualitative and quantitative effects of municipal waste landfills of the main urban settlements in Suceava County (Suceava City municipal landfill and Gura Humorului, Rădăuți, Siret, Câmpulung Moldovenesc, Fălticeni and Vatra Dornei urban waste landfills) on air quality. The dispersion of methane emitted from the largest MSW landfill in the county, the Suceava municipal landfill respectively, is also presented, taking into account seasonal, daytime and nighttime meteorological parameters.

KEY WORDS: methane emissions, carbon dioxide, pollution index, municipal solid waste, Gaussian modeling

1. Introduction

Biodegradable domestic waste landfills are important sources of air pollution by means of greenhouse gas emissions known as "landfill gas". Landfill gas generated can contain different components which are known to cause "greenhouse effects". Such components are the two main compounds of carbon: methane (CH_4) and carbon dioxide (CO_2).

Methane, reaching the atmosphere, is a shorter-life gas (7-12 years) compared to the carbon dioxide (100 years), but instead is 25 times stronger than carbon dioxide in what concerns the effect on climate change on a 100 years time scale.

The U.S. Environmental Protection Agency estimated that the total anthropogenic methane emissions for the whole world in 2000 equaled 282 mil. tones, of which approx. 13% were released from landfills (USA, Environmental Protection Agency, International analyses of methane emissions, 2002).

According to the estimates, the total annual amount of biological waste in the European Union is between 76.5 - 102mil. t. for food and gardening waste included in the mixed municipal solid waste, and 37 million tons for food and drink industry waste (ECC - Green Paper, 2008). Moreover, it is estimated that at European level the annual contribution of anthropogenic methane emissions is distributed as follows: waste disposal totals approx. 76.3%, underground mining - about 15.9%, intensive growing of poultry and pigs contributes with approx. 6.4% and treatment plants of urban waste water with approx. 1.4 %. Assuming that all countries should comply with Directive 1999/31/EC on waste disposal, even if there is an increase in the quantity of MSW, it is estimated that methane emissions in 2020, expressed as carbon dioxide equivalent, will be 10 million tones lower than in 2000 (IEC, Green Charter, 2008). Due to the increased contribution of methane, the second important "greenhouse effect gas" after CO₂, monitoring of landfill emission of this particular gas is required by the European Community Regulation EC 166/2006 (European pollutant release and transfer register "E-PRTR Regulation"). Under this Regulation, Member States are required to report annually the amount of methane released into the atmosphere by functional and closed landfills, along with the management of the landfill sites.

Landfill gas can be burned on site, whether to generate electricity or heat, or for decontamination and optimization in order to achieve the quality of fuel used for auto vehicles or of natural gas delivered to the national energy network, under Directive 2001/77/EC (RES Directive) on the promotion of electricity produced from renewable energy sources and Directive 2003/30/EC on biofuels.

Selection of options available for municipal solid waste management depends on a number of local factors, including: waste collection systems, waste composition and quality, climatic conditions, the potential use of various types of waste -derived products such as electricity, heat, gasses with high methane content or compost. These measures would contribute to the achievement of both the Kyoto targets on reducing greenhouse gas emissions and the objectives established for recoverable energy resources.

It is estimated that in Romania annual methane emissions of anthropogenic nature, for the recent years, amount to about 86000 tones (Ministry of Environment and Climate Change, National Strategy for Waste Management 2014-2020). The issue of waste management is topical for our country as well, which has undertaken to align to the Community provisions: to close the non-compliant landfills, to selectively collect waste and reduce the amount of waste disposed of in landfills (through recycling and treatment) and the content of biodegradable material contained in waste.

Referring to Suceava County, currently none of the 7 MSW landfills, classified by types of waste disposed of as non-hazardous locations (Class "b"), is not in accordance with the provisions of the Directive 1999/31/EC on waste disposal. These have adverse effects not only on the landscape, soil and groundwater quality, but also on air composition, due to high emissions of greenhouse effect gases.

2. Physico-chemical mechanisms of landfill gas production and their effects on air quality

Landfill gas generated by biodegradable household waste can contain different components with greenhouse gas effects. Such components are mainly represented by two carbon compounds: methane (CH₄) and carbon dioxide (CO₂), representing over 90% of landfill gas. Other compounds produced by MSW landfills have a much lower share: about 2-3% of landfill gas is represented by

mercaptans and the rest, by hydrogen sulfide, ammonia and so on, giving the well-known odor characteristic of landfills and being able to affect the health of exposed persons (Chiriac et al., 2007).

Multiphase nature of municipal waste and the significant influence of biological factors on waste degradation cause great complexity in biochemical processes. Thus, it is possible to distinguish several steps in the biodegradation process of waste: degradation of organic polymers to sugars (by hydrolysis), their transformation into acetic acid (acidogenesis) and the generation of CH₄ (methanogenesis), the main component of landfill gas. These processes are primarily anaerobic, with the exception of the initial phase, when oxygen is still available, thus enabling degradation. Another phase is the oxidation of CH₄ in the upper layers of the landfill. At this level, interface with the atmosphere increases the amount of available oxygen and allows the development of methanotrophic microorganisms that oxidize CH₄ to CO₂, thus reducing environmental impact, given that CO_2 is a less dangerous greenhouse gas than CH₄ (Czepiel et al., 1993).

Within the layers of solid municipal waste, concentration of toxic components that cross waste layers depends on the quantity of waste, the rate of mass formation/degradation of intermediates (hydrolysis and acidogenesis), the mass emission rate of the final products (mass transfer) and the process of dispersion in the atmosphere.

According to EC Regulation 166/2006 on the quantities of pollutants released and transferred to the environment (E- PRTR Regulation) the threshold value for CH_4 released by a source into the atmosphere is 100000kg/yr. CH_4 is not standardized for immissions for the protection of human health and vegetation, being considered that it has no immediate toxic effect (only at very high concentrations of over 1500000µg/m³, CH_4 may adversely affect human health - Cotrau et al., 1991).

3. Data and methods used

The paper is based on the following information:

- information on MSW generation (types of solid waste generated and collected), provided by sanitation operators and administrators OF landfills, which is based on estimates and not on precise data obtained by weighing;
- calculation of methane emissions from active MSW disposal, using the calculation method described in the document IPCC Good Practice Guidance and Uncertainty Management in National Greenhouse Gas Inventories;
- calculation of methane emissions after closure of municipal landfills over a given period, using the calculation method described in "Revised 1996 IPCC Guidelines for National Greenhouse Gas Inventory, Vol. 3 - Reference Manual, Ch. 6 - Waste";
- calculation of pollution indices for methane emissions, in order to evaluate the size of the impact on air quality, with the purpose of implementing practices of waste capture, treatment and recovery as energy resource;
- averaging of multiannual hourly values of the meteorological parameters (wind speed and direction, cloud cover, global solar radiation intensity), for seasons, daytime and nighttime;
- averaging of hourly values of incident solar radiation for daytime in 2012;
- calculation of dispersion of methane released in 2012 by the largest solid waste landfill in the county, the Suceava MSW landfill respectively, using a Gaussian model to estimate long-term average concentrations of pollutants for surface sources.

4. The calculation method of methane emissions

Calculation method of methane emissions for the amounts of waste disposed of on functional landfills

In order to calculate methane emissions in Tab. 1, resulted from the disposal of municipal solid waste on landfills, we used the method described in the document "IPCC Good Practice Guidance and Uncertainty Management in National Greenhouse Gas Inventories", Chapter 5 - Waste, and for determining the amount of methane released in the year (t) we used [2] described in "Default Method - Tier I" as follows:

$CH_{4 (Gg/an)} = [(MSW_T \times MSW_F \times L_o) - R] \times (1 - OX)$ [2] where:

MSW_T - the total amount of MSW generated (Gg/year), 1Gg = 1000 tonnes,

MSW_F - fraction of MSW removed at the disposal site,

 L_0 - methane generation potential, depending on the morphological composition of waste and calculated according to [3],

R - methane recovered in the inventory year t, (it is recommended that R = 0, assuming that methane is not collected in order to be flame burnt),

OX - oxidation factor (it is recommended that OX = 0 for the undeveloped landfills, or OX = 0.1 for the well-developed landfills).

The methane generation potential is given by the formula:

$L_{o (Gg C/Gg DMS)} = [MCF \times DOC \times DOC_F \times F \times 16/12]$ where: [3]

MCF - Methane correction factor, for which the corresponding value in the column "Methane correction factor (MCF) Default values" from Table 1 should be chosen as follows:

able 1. The values established for methane correction factor (MCF) – according to the landfill type							
Landfill type	MCF Value						
- administrated	1,0						
 not administrated - deep(≥ 5m of waste) 	0,8						
 not administrated - less deep (≤ 5m of waste) 							
- undefined	0,6						

DOC - degradable organic carbon (Gg C/Gg MSW), for which [4] is used, and the result is divided by 100:

DOC_f - dissimilated DOC fraction = 0.55 (in the range 0.5 to 0.6);

F - methane fraction in biogas (by volume), using nationally recommended value of 0.5; 16/12 - coefficient of carbon (C), conversion into methane (CH₄).

DOC
$$_{(GgC/GgDMS)} = (0,4 \times A) + (0,17 \times B) + (0,15 \times C) + (0,3 \times D)$$
 [4] where:

A - the MSW fraction represented by paper and textiles;

B - the MSW fraction represented by garden waste, parks waste and other biodegradable organic waste (except food waste);

C - the MSW fraction represented by food waste;

D - the MSW fraction represented by wood and straw waste.

Calculation of methane emissions from closed landfills

For the same waste landfills, in the post - closure period for calculating methane emissions (Table 1) we used the calculation method described in the document "Revised 1996 IPCC Guidelines for National Greenhouse Gas Inventory, Vol. 3 - Reference Manual, Chapter. 6 - Waste", which presents the estimation model of the evolution of methane emissions over a period of time equal to a decade ([5]):

 $Q = L_0 R (e^{-kc} - e^{-kt})$ [5] where:

Q - volume of methane generated in the current year (m³/year);

Lo - methane generation potential (m³/Mg waste) 1Mg = 1tonne;

R - the average annual rate of accepted waste to be disposed of on the landfill, during the functional period of the landfill (Mg/year);

k - Methane generation constant rate (1/year),

c - time elapsed from landfill closure (number of years);

t - time elapsed since the opening of the landfill (number of years);

e = 2.71828 (Euler's constant).

Methane generation potential L_0 depends on waste composition, especially on the cellulose content, which varies between 100-200m³/Mg according to the results shown by studies.

The average annual rate of accepted waste to be disposed of on the landfill, during the functional period of the landfill (R) is calculated as follows: the sum of the amounts of waste disposed of each year throughout the whole functional period of the landfill, divided by the number of years of its operation.

Methane generation constant rate (k) is dependent on the environment in which the landfill is located. Higher levels are associated with high humidity, but these are influenced by pH, temperature and other environmental factors. This constant can vary from less than 0.005 to 0.4 (1/year). The recommended values for k, according to the AP - 42 methodology (USA - EPA, 1991) are: k = 0.02 for areas with rainfall < 25inch/yr (i.e. 635 mm/year, 1 inch = 25.4mm) and k = 0.04 for areas with rainfall > 25inch/yr (635mm/year). The implicit pair (L₀, k), recommended by the AP -42 methodology is 100m³/tonnes of waste for both k = 0.04 (for landfills located in wet environment) and k = 0.02 (for landfills located in dry environment).

Since the average rainfall in Romania amounts to 637 mm/yr, with large amounts characteristic of mountain areas (over 1000 mm/yr) and lower in Baragan (under 400mm/yr), it is recommended that the following values be used: $L_0 = 100m^3/yr$ and k = 0.02 (for landfills located in dry environment).

Time elapsed since landfill closure (c) equals the number of years passed since the landfill was closed (without including the year of the closure) until the year in which the calculation of methane emissions is performed.

Time elapsed since landfill opening (t) equals the number of years which represent the functional lifetime of the landfill.

To convert the volume of methane Q (m^3 /year) released from waste landfills into methane amount (kg /year), [6] is applied:

Methane quantity (CH₄) (kg/year) = Q (m^3 /year) × methane density [6]

For the density of methane released from waste the value 0.73kg/m³ was considered, based on a study performed by German and Romanian experts (Blasy et al., 2006).

Characteristics of MSW landfills in Suceava County and estimation of the pollution index

Table 2. Data on urban waste landfills and CH₄ emissions in Suceava – Source: Environmental Protection Agency Suceava

Ref. No.	Landfill	Area (ha)	Opening year	Day, month, year of closure	Year	Amount of waste (tonnes)	Amount of methane released (kg/yr)	Threshold val. according to R. CE 166/2006 (kg/yr CH4)	PI % methane
1	Suceava	11.6	1072	31 12 2008	2007	102220	3360000	100000	-94,2
1	Suceava	11.0	1572	51.12.2008	2008	135298	2097872	100000	-90,9
					2009*	1400000	1400839	100000	-86,7
Current					2010*	1200000	1145134	100000	-83,9
Sucea	va post-closure				2011*	1000000	910767	100000	-80,2
					2012*	800000	695873	100000	-74,9
	Rădăuți				2007	10079	259726	100000	-44,4
2		4.435	1984	31.12.2009	2008	11486	285007	100000	-48,1
					2009	18184	469477	100000	-64,9
					2010	200000	218182	100000	-35,9
Rădău	ıţi² - post-closure				2011	200000	206887	100000	-34,8
					2012	200000	195777	100000	-32,4
					2007	12297	283223	100000	-47,8
2	Fălticeni	ălticeni 1.0 1978	1079	31.07.2010	2008	19694	399296	100000	-59,9
3			1978		2009	53933	924801	100000	-80,5
					2010	31724	634120	100000	-72,8
Fältic	ani ³ nost closure				2011	117600	121474	100000	-9,7
Fairle	eni - post-ciosure				2012	117600	116296	100000	-7,5

¹ The Suceava MSW landfill was closed in 2008. Until 2008 (inclusively), **Tab. 2** operates with annual quantities of waste disposed in this particular landfill and correspondent annual emissions of CH_4 , but from 2009 it operates with total annual amounts of existing waste for the entire landfill and with total annual CH_4 emissions from the entire mass of waste.

²The Rădăuți MSW landfill was closed in 2009. Until 2009 (inclusively), **Tab. 2** operates with annual quantities of waste disposed there and correspondent annual emissions of CH₄, and from 2010 it operates with total annual amounts of existing waste for the entire landfill and with total annual CH₄ emissions from the entire mass of waste.

^{3, 6, 7}For the MSW landfills of Fälticeni, Gura Humorului and Campulung Moldovenesc towns, the annual amounts of waste disposed, the annual amounts of CH₄ released by these landfills, total amounts of waste per year for all landfills and annual quantities of CH₄ released by landfills in different years were measured and calculated similar to the previous cases: Suceava and Radauti.

					2007	8436	135000	100000	-14,9		
				31.12.2011	2008	8732	153106	100000	-20,9		
4	Gura Humorului	2.126	1955		2009	9109	154952	100000	-21,5		
					2010	25650	443378	100000	-63,2		
					2011	22069	357524	100000	-56,3		
Gura I	Humorului ⁴ - post	-closure			2012	137080	116851	100000	-7,8		
		1.62		31.12.2011	2007	18814	224170	100000	-38,3		
					2008	10486	183106	100000	-29,4		
5	Câmpulung Moldovenesc		1990		2009	17764	316560	100000	-52,0		
					2010	24533	446664	100000	-63,4		
					2011	24557	437754	100000	-62,8		
Câmp	ulung Moldovene	sc⁵ - post-	closure		2012	102845	115529	100000	-7,2		
c		0.0 40	1070		2007	5115	90800	100000	4,8		
6	Siret	0.8 197	1970	31.12.2008	2008	4425	83820	100000	8,8		
Siret -	Siret - post-closure					2009 Covered by land and inert waste					
7	Vatra Dornei	1.7	1983	16.07.2005	Covered by land and vegetation						

Table 2 shows the characteristics of MSW landfills in Suceava County: area, the amounts of waste disposed of starting with 2007 and until closure, the amounts of methane emitted into the atmosphere, both during the functional period and post-closure periods, including the year 2012.

Moreover, pollution index (PI) for methane emissions is rendered, compared to the threshold value required by Regulation EC 166/2006.

The average pollution indices, calculated for methane released from the Suceava County landfills, are shown in Figs 1 and 2.



Figure 1. PI% calculated based on averaging the annual amounts of waste disposed before the closure of landfills.

Figure 2. PI%, calculated based on the total amount of waste disposed on the surface of landfill platform areas (post-closure).

From the data presented in Tab. 1 and 2, and in Figs 1 and 2 respectively, the following instances on methane emissions since 2007 can be observed:

Obs. * In 2009, 2010, 2011 and 2012 for the Suceava landfill, 2.0ha of the landfill area were covered annually with inert waste from construction (rubble, construction earth, rocks, earth and vegetal waste from parks etc.).

Suceava MSW landfill:

- for the 2007-2008 functional interval mentioned in the paper, the threshold value imposed by *Regulation EC 166/2006* of 100000kg/yr methane was exceeded 21-33 times, and during the post-closure period 2008-2012, the threshold value was exceeded 6 to 14 times;
- the average pollution indices for the functional period (90.2%) and post closure period (83.6%)
 place this particular pollution source in the "very bad" pollution level (significant pollution with
 destructive effects on the environment, decontamination measures being needed urgently).

Radauti MSW landfill:

- for the functional interval 2007-2009, the annual amounts of methane released into the air exceeded 2.5 to 4.7 times the threshold value, and during the post-closure period 2010-2012 the exceedance was about two times higher than the threshold value;
- the average pollution indices during landfill utilization (52.4%) and for the post-closure period (34.8%) place the landfill site in the "very bad" pollution level.

Falticeni MSW landfill:

- during the functional interval 2007-2010, the annual amount of methane released into the air exceeded 2.8 to 9.2 times the threshold value and during the post-closure period 2011-2012, the exceedance was about 1.2 times higher than the threshold value;
- average pollution index of 65.2%, characteristic of the functional period, placed the landfill in the "very bad" pollution level, whereas the 8.6% PI of the post-closure period placed the landfill in the "bad" pollution level.

Gura Humorului urban solid waste landfill:

- between 2007 and 2011, when the landfill was functional, the annual amount of methane released into the atmosphere exceeded 1.3 to 4.4 times the threshold value, and in 2012, after the cessation of landfill activity, the exceedance was about 1.16 times higher than the threshold value;
- average pollution index (19.1%) during landfilling activities placed the landfill in the "very bad" pollution level, whereas the 7.8% PI in the post -closure period is characteristic of "bad" pollution level.

Campulung Moldovenesc MSW landfill:

- during the activity period (2007-2011), the annual amount of methane released into the air exceeded 1.8 to 4.5 times the threshold value, while in the post-closure period 2010-2012 the exceedance was 1.2 times the threshold value;
- average pollution indices during the activity period (39.9%) and post -closure period (7.2%) are characteristic of the "very bad" pollution level.

For Siret urban solid waste landfill, during the 2007-2008 landfilling period, the annual amounts of methane emitted into the air did not exceed the threshold value and the 6.6% pollution index places the landfill in the "medium" pollution level (significant pollution with possible manifestations of pollution phenomenon).

For Vatra Dornei landfill methane emissions were not calculated, the landfill being closed in 2005 and therefore not covered by *Regulation EC 166/2006*.

In conclusion, the worst situation is characteristic of Suceava MSW landfill because of its large area which totals 11.6ha and significant amounts of "landfill gas" released into the air, especially methane.

5. Calculation of dispersion of methane released from the Suceava MSW landfill

Mathematical models for calculating concentrations of pollutants in atmosphere describe pollution mechanisms in order to estimate the impact of pollutants on the environment. Dispersion models are very useful tools in many situations where data from direct measurement are not available or existing data are inadequate (Sandu et al., 2004).

The models most frequently used to calculate pollutant concentrations are based on Gaussian solutions of the diffusion equation (Pasquill and Smith, 1983).

Pollutants dispersion into the atmosphere depends on a number of factors which act simultaneously, namely (Pasquill, 1983; Tumanov, 1989; Turner, 1994): the factors characterizing emission sources, meteorological factors specific of the environment in which the pollutant emission occurs and which determine the horizontal and vertical dispersion of pollutants, factors that characterize the area where the emission occurs, as well as atmosphere response.

For methane released by Suceava MSW landfill (Fig. 3), the dispersion into the atmosphere was determined by using an ISC model for estimating long-term averages of pollutant concentrations for surface sources (ISCLT3 - Industrial Source complex - Long Term).



Figure 3. Suceava MSW landfill (source: Google Earth); the inset - left corner, the same landfill (photo Environmental Protection Agency Suceava).

Mathematical modeling allows for:

- Establishment of a network of air quality theoretical monitoring by calculating maximum concentration on 8 wind directions, at different distances from the landfill site (from 300 to 5000m, for each 100m up to 3000m and for each 500m between 3000 and 5000m);

- Calculating methane concentrations taking into account wind speed and thermal stability classes (after Pasquill, 1983) for nighttime and daytime during each season - Table 4;

- Calculation of mixing height (H_{mixing});

- Calculation of average concentrations at soil level on the long term (night/day, seasonally) using average seasonal multiannual wind frequencies for 8 directions.

Meteorological parameters for Suceava City used in modeling. The data necessary to run the program, shown in Fig. 3 and Fig. 4, were obtained as follows: Solar radiation intensity was calculated by computing the hourly average values for 2012, as well as data from the Suceava automatic air quality monitoring station (SV1). Average seasonal cloud cover for daytime and nighttime was calculated from multiannual hourly values (2004-2012) for data recorded at Suceava Weather Station. Seasonal, daytime and nighttime wind speeds and frequencies were computed by processing multiannual hourly values for the same location and period.

Day and year interval	Cloud cover Daytime and nigh averages	ittime seasonal	Solar radiation intensity (W/m ²)		
	(10)	(8)	Seasonal daytime averages		
Winter nights	6,6	5,2	-		
Winter days	7,3	5,8	69,39		
Spring nights	6,3	5,0	-		
Spring days	6,9	5,5	238,67		
Summer nights	5,0	4,0	-		
Summer days	5,9	4,7	331,47		
Autumn nights	5,3	4,2	-		
Autumn days	6,2	4,9	176,47		

 Table. 3 Intensity of solar radiation (Source: SV 1 Station, EPA Suceava) and cloud cover (Source: Suceava Weather Station)

Pasquill (1983) defined six classes of atmospheric thermal stability, depending on wind speed and solar radiation (for daytime) and wind speed and cloud cover (nighttime) as follows: A - very unstable, B - unstable, C - slightly unstable, D - neutral, E - stable, F - very stable.

Turner (1994) classified incident solar radiation as "strong radiation"> $600W/m^2$, moderate radiation - between 600 and $300W/m^2$ and "low radiation"< $300W/m^2$.

Season										No
Nighttime /	Parameters	Ν	NE	E	SE	S	SV	v	NV	INO in al
Daytime										wind
Winter	Stability class	D	E	E	D	D	D	D	D	E / F
nights	H _{mixing} (m)	1056	-	-	1376	992	1152	1184	1472	-
Winter days	Stability class	С	С	С	С	С	С	С	D	E / F
	H _{mixing} (m)	1120	768	1024	1440	1056	1120	1152	1664	
Spring nights	Stability class	E	D	D	D	D	D	D	D	E/F
	H _{mixing} (m)	-	1248	1344	1632	1024	1056	992	1536	-
Spring days	Stability class	С	С	С	D	С	С	С	D	E / F
	H _{mixing} (m)	1312	1280	1024	1856	1280	1312	1184	1760	-

Table 4. Meteorological parameters - stability classes

	•									
Summer	Stability class	D	E	E	D	E	D	D	D	E/F
nights	H _{mixing} (m)	1216	-	-	1120	-	1120	1024	1312	-
Summer	Stability class	В	В	В	С	В	В	В	С	E/F
days	H _{mixing} (m)	1056	1024	1056	1440	1216	960	1088	1536	-
Autumn	Stability class	D	Е	E	D	E	D	D	D	E/F
nights	H _{mixing} (m)	1472	-	-	1056	-	992	1024	1344	-
Autumn	Stability class	С	С	С	С	С	С	С	С	E / F
days	H _{mixing} (m)	1120	992	960	1408	1024	1120	1248	1536	-

Multiannual data highlight for windy synoptic conditions prevailing wind direction from the northwest (minimum 25.3% for autumn nights, maximum 37.3% for summer days). Calm is dominant at night (the minimum equals 41.6% and is specific of winter nights, while the 58% maximum is characteristic of autumn nights).

The input data for methane dispersion modeling were:

- source type rural areas, surface source,
- mass emission rate (CH₄ emission rate) (g/s.m²) 0.00019 (calculated from Tab. 2 for 2012)
- landfill height 8m,
- landfill dimensions: length 449m, width 58m,
- distance of the observation point/receiver from the ground: 1.5m,
- Class of atmospheric thermal stability (A, B, C, D, E, F) according to Tab. 4,
- Wind speed (seasonally, for both nighttime and daytime in m/s) according to Fig. 4,
- Distances for methane dispersion modeling: minimum = 300m, maximum = 5000m.

Figs 5 to 20 show the results of data processing: methane concentration values at soil level for seasons, nighttime and daytime, for distances from 300 to 5000m, following wind direction.

Atmospheric calm, prevalent at night (in winter - 41.6%, spring - 49.4%, summer - 57.8%, autumn - 58%), is considered in the calculation of weighted average of concentration values according to **[8]**:

$$C_{mediu} = (\Sigma C_i^* f_i) / (\Sigma f_i)$$
 [8] where:

 C_i – concentration calculated for all wind types (µg/m³);

f_i - wind frequency (%).

In Figs 5 to 20 methane concentrations (μ g/m³) are presented, after having been automatically calculated for the four seasons of the year, for nighttime and daytime.

Methane dispersion in winter nights

During winter nights (Tables 3, 4 and Fig. 5), the average cloud cover is maximum reported to the annual profile for this diurnal range - 6.6 tenths; the predominant stability class is neutral, the average wind speed equals 3.2m/s, and the calm is dominant, with a percentage of 41.6% (Fig. 5 top left inset).

During calm intervals, pollutant mixture with air is less active, thus leading to an increase in methane concentration at the soil level, with maximum values of $5349\mu g/m^3$ (500m from the center of the MSW landfill), and minimum of $1207\mu g/m^3$ (5000m from the center of the polluting source). This distribution also applies to night and day calm intervals for other seasons (Figs 5 to 12, bottom left inset).

11



Figure 5. Methane dispersion for winter nights, wind speed and frequency roses for directions, methane isoconcentrations for inter-cardinal and cardinal directions at different distances from the center of Suceava MSW.

Analyzing the horizontal dispersion model of methane concentrations for synoptic windy conditions, we observe several relevant issues. NW winds, despite the highest frequency and speed (32.2% and 4.6m/s), fail to push pollutants southeastward due to land morphography and morphometry, which plays an important role in the distribution of pollutants not only in winter nights. Suceava MSW landfill is located on the right bank of the Suceava River, in the floodplain of the river, at an altitude of about 270m, on land widely open to NW, N, NE and E, but surrounded from SE, S, SW and W by a concave coastal scarp generally exposed to the north; between the base of the scarp and the top there is a difference of approximately 70m, plus 10 - 15m height of forest cover. The coastal scarp continues towards south and south -east with a long reverse, its elevation being gradually reduced in the directions mentioned. Once entering the concavity of the coastal scarp on which the landfill is located, much of the winds from the NW (which bring fresh air from the forests of northern and north- western sides of the city) are forced to climb the coast promontory and cross its reverse towards southwest. These winds undergo a slight Coanda effect, of redirectioning, caused by the presence of the northern extremity of the Cetății Hill on the western side of the landfill. Some winds are directed towards south or southweast. Towards northwest, polluted area falls under the influence of winds coming from this direction. An identical effect (the thinning out of the polluted area) is induced by winds blowing from the SE. NW winds, diverted towards southwest, lengthen significantly in this direction the polluted area of methane emissions.

The SW winds (F = 10.6%, v = 3.6m/s) limit the dispersion in this direction, pushing some of the pollutants towards NE (direction for which winds have low frequency and speed (0.6% and 2,9m/s respectively). Phenomenologies and similar distributive situations are also typical of nights in other seasons.

Methane dispersion in spring nights

For spring nights, atmospheric calm has a share of 49.4% (Fig. 6 - top left inset). With regard to the distribution of methane concentration we also referred to winter nights. Analyzing horizontal dispersion of methane for windy conditions - Fig. 6, (provided that cloud cover is 6.3 tenths, neutral stable conditions prevail, average wind speed is 3.8m/s, the mixing height is located approximately at 1262m, wind frequency and speed roses for directions are similar to those for winter nights, except for slightly higher wind speed), we find that this is very similar to methane concentrations dispersion during winter nights - Fig.5.



Figure 6. Methane dispersion for spring nights, wind speed and frequency roses for directions, methane isoconcentrations for cardinal and inter-cardinal directions at different distances from the center of Suceava MSW.

Winds from the NW diffuse methane more southerly, an important role in the dispersion being played by the concave shape of the coast base and scarp where Suceava MSW landfill is located.

Methane dispersion in summer nights

For summer nights, atmospheric calm reaches its maximum frequency (57.8%). Methane isoconcentrations take in this case the shape of nearly concentric circles with their center placed on Suceava MSV landfill. 500m from the circle center, methane concentration reaches $5349\mu g/m^3$ and 5000m from the center $1207\mu g/m^3$. This distribution also applies to atmospheric calm intervals during day and night for all seasons, and therefore, we will not insist on this distribution for such synoptic conditions.

In summer nights cloud cover and wind speed are small (5.0 tenths and 3.2m/s respectively). The most frequent stability classes are those neutral and stable, while mixing height is at 1158m. For windy conditions, methane distribution (Fig. 6) is very similar to that during winter nights (Fig.5). Prevailing winds from the NW (28.8%) stretch polluted area on this direction, dispersing pollutants downstream the Suceava River valley (towards SE, but mainly towards SV), as land morphology influences dispersion in this direction while also carrying methane to NE and N.



Figure 7. Methane dispersion for summer nights, wind speed and frequency roses for directions, methane isoconcentrations for cardinal and inter-cardinal directions at different distances from the center of Suceava MSW.

The dominant stability class is slightly unstable and mixing height is on average at 1168m. Winds from the NW (F = 37.3%, v = 5.2m/s), SW, S and SE have the highest frequencies and speeds. In windy conditions, under the influence of the other meteorological elements, local geomorphological factor (Suceava river valley axis orientation on NW-SE direction, the concavity of the coast scarp on the right bank of the Suceava River, where the river has developed its floodplain and where the Suceava MSW landfill is currently located, the wide opening towards NW, N, NE, E of Suceava floodplain and its remarkable smoothness etc.) characteristics of the polluting source, methane dispersion follows closely the SE and NV directions, but also SW and NE directions (Fig. 8).

Methane dispersion in autumn nights

In autumn nights atmospheric calm reaches 58 % (Fig. 8, top left), cloud cover is 5.3 tenths and dominant weather classes are neutral and stable. Mixing height is located on average at 1177m, with variations for certain directions.

Similar to summer nights, for atmospheric calm intervals, in winter nights methane concentrations reach a maximum (Fig. 8 bottom left inset). For windy conditions, under the influence of the NV dynamics (F = 25.3%, v = 4.2m/s), as well as S, SV dynamics and local topography, pollutant dispersion is mostly conducted on the SV and N directions (Fig. 8), very similar to the dispersion of methane in summer nights (Fig. 7).



Figure 8. Methane dispersion for autumn nights, wind speed and frequency roses for directions, methane isoconcentrations for cardinal and inter-cardinal directions at different distances from the center of Suceava MSW.



Figure 9. Methane dispersion for winter days, wind speed and frequency roses for directions, methane isoconcentrations for cardinal and inter-cardinal directions at different distances from the center of Suceava MSW.

Methane dispersion in winter days

In winter days air dynamics is activated, atmospheric calm having a share of only 27.5% of the time, while average wind speed is 3.7m/s in windy conditions. The intensity of solar radiation reaches averages daytime values of 63.39 W/m^2 , while cloud cover has the highest values (7.3 tenths, especially for stratiform cloud systems), and the days are the shortest of the year.

Methane dispersion in spring days

For spring days, air dynamics is most active. Atmospheric calm intervals reach the lowest values of the year (17.7%) and average wind speed reaches the highest values of the year (4.3m/s). On the background of a cloud cover of 6.8 tenths and with increasing day length, average daily values of solar radiation equal 238.67W/m². Prevalent weather class is slightly unstable and mixing height rises on average at 1376m (the highest of the year). Atmospheric circulation is dominated by NV winds (F = 36.6%, V = 5.5m/s) and SE winds (F = 15.1%, v = 5.8m/s), directions followed as well by methane released from Suceava MSW landfill (Fig. 9). The Suceava River Valley and local microlandforms have an important role in the spatial distribution of methane concentrations.



Figure 10. Methane dispersion for spring days, wind speed and frequency roses for directions, methane isoconcentrations for cardinal and inter-cardinal directions at different distances from the center of Suceava MSW.

Methane dispersion in summer days

For summer days, atmospheric calm has 23.9% of the time, and for these synoptic situations, spatial distribution of methane has been previously referred to. Average wind speed is 3.7m/s, cloud cover reaches 5.9 tenths and solar radiation intensity is maximum on an annual basis (331.47 W/m²). The unstable class is dominant and mixing height reaches 1172m. Dominance of NW winds

(F = 36.5%, v = 4.8m/s) and SE winds (F = 11.2%, v =4.5 m/s) clearly shape dispersion of methane released by Suceava MSW landfill along the valley axis (Fig. 10).



Figure 11. Methane dispersion for summer days, wind speed and frequency roses for directions, methane isoconcentrations for cardinal and inter-cardinal directions at different distances from the center of Suceava MSW.

Methane dispersion in autumn days

Autumn days have the highest interval of atmospheric calm of the year (31.1 %). When the wind blows, its average speed is 3.7m/s. Cloud cover specific to these days averages to 6.2 tenths and the intensity of solar radiation is $176.47W/m^2$.

Dominant stability class is slightly unstable and mixing height is located at 1176m. For the model obtained (Fig. 12) for different shares of the influence factors (climatic, geomorphological, polluting source characteristics), methane concentrations are spatially distributed in the form of concentric circles, which have at their center the landfill. Beyond similarities with diurnal distributions of methane for other seasons, the map of the spatial distribution of methane in autumn days (Fig. 12) shows features distinct from other similar maps (Fig. 9-11).

For weighted average values of methane concentrations in Fig. 5-12 (calculated according to formula 8), the analysis reveals several aspects. The highest values are recorded during nighttime for all seasons (Fig. 5-8), compared with those of daytime (Fig. 9-12). The highest values are specific to summer nights (Fig. 7) and spring nights (Fig. 6) when atmospheric calm has the highest frequency. The lowest values of methane concentration are recorded during spring and summer days (when atmospheric calm frequencies are low - 17.7% - Fig. 10 and 23.9% - Fig. 11 respectively, bottom left).



Figure 12. Methane dispersion for autumn days, wind speed and frequency roses for directions, methane isoconcentrations for cardinal and inter-cardinal directions at different distances from the center of Suceava MSW.

5. Conclusions

Currently, the management of Suceava County MSW landfills faces a number of problems concerning the protection of air quality:

- Urban waste disposal is carried out on bare land;
- Most current MSW landfills are not properly operated to prevent emissions of greenhouse gas, spread of fire, smoke and odors from waste auto ignition;
- there are no facilities to recover biogas produced by landfills;
- methane emissions for Radauti, Fălticeni, Gura Humorului, Campulung Moldovenesc landfills exceed by 2 to 5 times the threshold required by Regulation 166/2006 of the European Commission;
- methane emissions from Suceava MSW landfill platform currently exceed 16 times the threshold required by Regulation 166/2006 of the European Commission;
- For most urban solid waste landfills in Suceava County, concerning methane emitted into the atmosphere, pollution indices fall within the "bad" and "very bad" pollution levels. It is thus necessary implementation of measures for ecological reconstruction in those areas, biogas uptake and its use for heat generation (especially for the Suceava MSW landfill).

Through the implementation of mathematical modeling of methane dispersion in the MSW landfill area, the following has been observed:

- ✓ due to high frequencies of atmospheric calm (eg. the average frequency of 58.0% in autumn at night), the maximum methane concentrations exceed the 5000µg/m³ threshold at 500m from the polluting source and the 1200µg/m³ threshold at 5000m distance from the source;
- ✓ methane released by Suceava MSW landfill has maximum averaged values equal to 3543µg/m3 at 500m from the source and 736µg/m³ at 5000m during winter nights;
- ✓ for the prevailing NW wind direction (with frequencies between 25.3 and 37.3%) due to high speeds (ranging from 4.1 to 5.5m/s) and high mixing heights (between 1312m and 1760m) horizontal dispersion is favored (towards SE for days of winter, spring and summer; towards southwest and to a lesser extent towards S and SE for the nights of all seasons), as well as vertical dispersion;
- ✓ winds from the SW and SE are second as importance in pollutant dispersion towards N and NW;
- ✓ frequently, similar to Suceava situation, local geomorphological factors disrupt to a great extent pollutant dispersion; consequently, aerodynamic studies and simulations should play a more important role in setting the location of future landfills.

During winter, summer and autumn nights, methane pollution spreads from Suceava MSW landfill to residential areas in the city center, George Enescu and partly Obcini and Zamca areas. On spring, summer and autumn nights, pollution spreads northward over a large part of the Burdujeni area. During spring nights methane pollution strongly affects Lisaura locality.

During winter, spring and autumn days, pollution dispersion follows a northwest and southeast direction (along the axis of the Suceava River valley) and affects only to a lesser extent Suceava city residential areas, but partially affects the airspace of Lisaura locality. In autumn days methane polluted area becomes almost circular in shape (having the MSW landfill in the center), affecting the north-western suburbs of Suceava and north- eastern side of Lisaura locality.

Acknowledgements

Bistricean Petrut-Ionel would like to specify that this paper has been financially supported within the project entitled "SOCERT. Knowledge society, dynamism through research", contract number POSDRU/159/1.5/S/132406. This project is co-financed by the European Social Fund through the Sectoral Operational Programme for Human Resources Development 2007-2013. Investing in people!"

References

Blasy L., Lange M., Hagen N., Rosal D., Atudorei A. 2006. Beneficiile utilizării gazului de depozit rezultat din depozitul de deșeuri municipal, *Rev. Salubritate* 4 (20).

Cojocaru I. 1995. Surse, procese și produse de poluare, p. 178-186. Editura Junimea, Iași.

Cotrău M, Lidia Popa, Stan T., Preda N., Kinsses-Ajtay M. 1991. *Toxicologie*, pp. 106. Ed. Didactică și Pedagogică, Bucuresti.

- Czepiel P.M., Crill P.M., Harriss R.H. 1993. Methane emissions from municipal wastewater treatment processes, *Environ. Sci. Technol.*
- Diţoiu Valeria, Holban N. 2005. *Modificări antropice ale mediului,* p. 115-140. Ed. Orizonturi Universitare, Timişoara.
- Macoveanu M. 2003. *Metode și tehnici de evaluare a impactului ecologic*, p. 126-139. Editura Ecozone, Iași.
- Pasquill F., Smith F.B. 1983. Atmospheric Diffusion, 437 pp, Third Edition. Ellis Horwood Ltd, Chichester, England.
- Sandu I., Pescariu I.V, Sandu I. 2004. *Modele de evaluare a dispersiei poluanților în atmosferă, p*. 50-72. Ed. Sitech, Craiova.
- Tumanov S. 1989. Calitatea aerului, p. 9-38, 64-70. Ed. Tehnică, București.
- Turner D.B. 1994. Manual de estimări ale dispersiei atmosferice. Editura Lewis.
- ***Anuare privind starea mediului 2005-2012, A. P. M. Suceava.
- ***European Communities Commission, (2008), Green Paper the management of bio-waste in the European Union, SEC2936.
- ***Regulation no. 2008/98/EC on waste.
- ***Regulation no. 1999/31/EC on waste disposa.l
- ***Regulation 2001/77/EC –RES Regulation on promotion of electricity produced from renewable energy sources.
- ***Regulation 2003/30/EC on biofuels.
- ***Guidance Document for the implementation of the European Pollutant Release and Transfer Register E-PRTR, European Commission, Releases to air, 2006.
- ***H.G. nr. 349/2005, privind depozitarea deşeurilor.
- ***ISCLT3 Industrial Source Complex Long Term http://www.lakes-environmental.com/ lakeepa3.html.
- ***IPCC- Good Practice Guidance and Uncertainty Management in National Greenhouse Gas Inventories, Default Method – Tier I.
- ***Ministerul Mediului și Schimbărilor Climatice, Strategia Națională pentru Gestionarea deseurilor 2014-2020.
- ***Monitorizarea calităţii atmosferei Elemente metodologice privind elaborarea inventarelor de emisii şi modelarea dispersiei poluanţilor atmosferici Vol. I, Manual elaborat în cadrul activităţii de «Întărirea capacităţii de Monitorizare a Calităţii Aerului în România", 1997.
- ***Plan Județean de Gestiune a Deșeurilor în județul Suceava.
- ***Revised IPCC, Guidelines for National Grenhouse Gas Inventory, Vol. 3-Reference Manual, Cap. 6-Waste, 1996.
- ***Regulation (EC) No 166/2006 of the European Parliament and of the Council concerning the establishment of a European Pollutant Release and Transfer Register ("E-PRTR Regulation"), Anexa 1 Activities, 5d, Anexa 2- Pollutants, Methane (CH4).
- ***USA, Environmental Protection Agency, (2002), International analyses of methane emissions; www.epa.gov/methane/intlanalyses.html.
- ***USA, Environmental Protection Agency, 1991, Metodologia AP-42.