### METHODS TO ASSESS THE IMPACT OF POLLUTANTS DISCHARGED INTO THE ATMOSPHERE BY MANUFACTURERS OF REFRACTORY MATERIALS

Radu Vasile BUZDUGA<sup>1</sup>, Miron Dorica BUZDUGA<sup>2</sup> Adriana Mioara COMSA<sup>3</sup>, Aurica GOLEANU<sup>4</sup>

The refractories industry is cited as a polluting industry. The most important agents encountered in this area are dust, gases, waste resulting from the processes. In this paper we present current methods used to assess the impact of pollutants discharged into the atmosphere by the producers of refractory materials, highlighting the practical application of these methods by HELIOS Astileu, Romania

**Keywords**: pollutants, atmosphere, refractory materials

#### 1. Introduction

Refractory industry differs significantly from other sectors of the ceramic industry. The field of silico-aluminous refractory studied in this project is related to some extent to the one of building bricks for the flow of mixture-compression, combustion, as well as the production of cement, in particular in the preparation of firebricks.

Silico-alumina refractory obtaining involves the use of raw materials with high content of free silica, extremely harmful to human health. In this paper we present current methods used to assess the impact of pollutants discharged into the atmosphere by the producers of refractory materials, highlighting the practical application of these methods by HELIOS Astileu.

The Gaussian air pollutant dispersion equation (discussed above) requires the input of H which is the pollutant plume's centerline height above ground level—and H is the sum of Hs (the actual physical height of the pollutant plume's emission source point) plus  $\Delta H$  (the plume rise due the plume's buoyancy).[1][15]

<sup>2</sup> PhD eng, Research and Development Institute for Refractories and Ceramic Products – CCPPR Alba Iulia, Romania, e-mail: ccppr@rdslink.ro

<sup>&</sup>lt;sup>1</sup> PhD eng.., Research and Development Institute for Refractories and Ceramic Products – CCPPR Alba Iulia, Romania, e-mail: radu buzduga@yahoo.com

<sup>&</sup>lt;sup>3</sup> Eng., Research and Development Institute for Refractories and Ceramic Products – CCPPR Alba Iulia, Romania, e-mail: e-mail: ccppr@rdslink.ro

<sup>&</sup>lt;sup>4</sup> Eng., Research and Development Institute for Refractories and Ceramic Products – CCPPR Alba Iulia, Romania, e-mail: ccppr@rdslink.ro

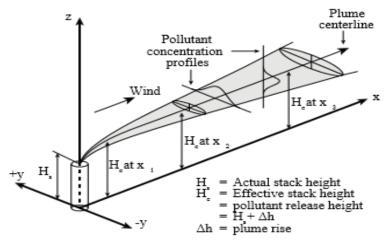


Fig. 1. Visualization of a buoyant Gaussian air pollutant dispersion plume, [1]

To determine ΔH, many if not most of the air dispersion models developed between the late 1960's and the early 2000's used what are known as "the Briggs equations." G.A. Briggs first published his plume rise observations and comparisons in 1965. In 1968, at a symposium sponsored by CONCAWE (a Dutch organization), he compared many of the plume rise models then available in the literature.[2] In that same year, Briggs also wrote the section of the publication edited by Slade dealing with the comparative analyses of plume rise models.[3] That was followed in 1969 by his classical critical review of the entire plume rise literature, in which he proposed a set of plume rise equations which have become widely known as "the Briggs equations". Subsequently, Briggs modified his 1969 plume rise equations in 1971 and in 1972.[4], [13]

### 2. Experimental work

### 2.1. Overview of pollutant emissions from

The most important component of pollution in this field is the dedusting one. Particulate emissions and immissions from technological processes for manufacturing refractory products is very important because they contain free crystalline silica and heavy metals.

The effects of HELIOS Astileu activity on the environment will be estimated according to the pollutants emitted and affecting environmental factors.

The recoverable products of this company are accompanied by gaseous, solid and liquid emissions.

Reduction or elimination of these emissions is imposed by environmental legislation and is the basic condition for further processes, the solution currently practiced is generally to discharge to the environment of pollutants and quantities of raw materials.

The company is located in the north-east of the country, in Bihor county, 35 km from the city of Oradea, in the east of the village Astileu, south-east of the town Alesd.

The dump of industrial wastes of the company is situated at a distance of 500 m from the boundary to the town Alesd, along Crisul Repede valley in the eastern part of the village Astileu.

The total area of the dump is 27.839 sqm. The dump has a length of 1,150 m and a height of 8.5 m with an average slope of 60 degrees. The large base of the dump is approximately 18 m, and small base is approximately 12 m. The volume of stored waste is approx. 150.000 m<sup>3</sup>.

To assess the impact of pollutants discharged into the atmosphere two methods were used, namely:

- Climatological model of pollutants dispersion,
- Bosanquet-Peason mathematical model.

## 2.2. Presentation of the method of calculating the dispersion of pollutants using climatological dispersion model.

The matematical model used to evaluate the impact of evacuated pollutants in the atmosphere is the climatologic model SIMPG V3 for calculation of concentrations field.

Using this model was made taking into account that it allows the simultaneous intake of several sources to total concentration in a receiver; allows estimation of short term and long term; is applicable to continuous point and surface sources.

The fundamental physical basis of the model is the assumption that the spatial distribution of concentrations is given by the Gaussian plume pollutant. The model is therefore a Gaussian model combined with the mediation of the concentration on long intervals.[14], [17]

Gaussian solving of the diffusion equation has proved suitable for dispersion modeling. Complex experiments conducted by research groups in the world and in Romania showed a good degree of matching theoretical and experimental data: in addition it occurs advantage of facilities related to the development of computer programs, making the model an operational tool. Gaussian models are three-dimensional models that allow calculation of pollutant concentrations at a certain height (including the ground level) and at a certain distance from the source. The input data include information on computing grid, data broadcasting and meteorological parameters. The output data of the model is calculating every point grid covering the area of influence of the sources of the average concentration of seasonal or annual reports of each pollutant, the maximum concentration in 30 min. (The highest value involving all weather situations possible) and seasonal or annual average frequency of achieving and

exceeding a threshold concentration per 30 min. (sanitary standards, the limit value for the protection of vegetation, etc.).

Based on these data we can trace on the area map the curves of iso-concentrations and iso-frequencies that emphasize spatial distribution of concentrations area and the level of atmospheric pollution and the long-term and short-term exposure. It is to be mentioned that if a group analysis of the impact of sources, measurements calculated in each grid point totals contributions of all sources. Using the presented climatological model the average short-term concentrations were calculated, daily and yearly for pollution sources within the company Helios Astileu. Data input into the program were taken from the previous tables that presented physical characteristics of sources, emission rate, flow and speed of the exhaust gases into the atmosphere. Dispersion calculations were made under the following conditions: simultaneous emission of all sources, disadvantageous conditions climatological, just to get the maximum concentrations that may be recorded.

There have been calculated dispersions for pollutants whose emissions have been assessed as the greatest ( $SO_2$  and total dust). The area for wich the concentrations of discarded pollutants were calculated comprises the receptors in the area.[5]

### 2.3. Presentation of method of calculation for the dispersion of pollutants using the mathematical model Bosanquet-Pearson.

The dispersion of pollutants was compiled based on measurements from sources based on material balance and on the fixed measurements.

The factors that influence the dispersion of pollutants in the atmosphere are factors that characterize the source (stack height exhaust temperature, speed, quantity emitted), the relief and roughness of the zone in which the dispersion occurs, factors characterizing the air space in which the emission occurs (meteorological factors).

Pollutant dispersion process can be studied through mathematical modeling, allowing expression by calculating the spatial distribution in case of pollutants studied.

Starting from the equation of diffusion of pollutants in a system of interaction and introducing a number of simplifying assumptions, we reach to a solution as a Gaussian distribution.

Among the simplifying assumptions the most important are the emission source which is punctual, the emission source is located on relatively flat terrain, the turbulence is homogeneous and stationary throughout the air mass, the axis 0x is oriented downwind and the pollutants that are conservative (not undergoing physical or chemical changes).

Qm mass flow rates of pollutants (mg/s) entering the dispersion calculation are determined directly by measuring the concentrations of pollutants at the source in the exhaust gases into the atmosphere, or indirectly through assessments on the technological balance of materials.

$$Qm = Cc * Qv \tag{1}$$

, where: Cc - is the concentration of the pollutant and Qv - is the volumetric flow rate of the source.

The wind interferes into the dispersion through velocity and direction.

The calculation of wind speed at a certain level is usually based on ground wind velocity (standard height  $H_I = 10$  m) with a relation of the form:

$$V = Vo(H/H1) \tag{2}$$

, where: V = average wind speed at the height H, m/s

 $V_o$  = average wind speed at the reference level  $H_1$  in m/s;

 $H_1$  = effective height clearance in m;

$$\Delta h = h + h1 \tag{3}$$

, where h = geometric height of the source,  $m \Delta h = \text{heightening}$ ; N turbulence index.

Values of n depend upon atmospheric conditions.

Wind direction sets the travel of the mass of pollutants. The speed determines the direction of movement of the pollutant on heightening influence. This is defined as the distance that travels up from the exit from the chimney to the initial loss of momentum. This distance is inversely proportional to wind speed.

Heightening wedge pollutant was calculated using the following calculation equation:

$$\Delta h = 1.9 D U/V \tag{2}$$

, where: V = average wind speed at the height H, m/s;D - represents the inner diameter of the exit at the chimney mouth, m; U - gas speed in the chimney section in m/s.

Based on the mathematical model an algorithm was developed that, in a certain meteorological situation, you can calculate the maximum concentration of the pollutant and distance from the source: you can also perform calculations of concentration on an axis with a step by choice for all stability classes and wind speeds compatible.

Dispersion calculations were made for pollutants for which recorded values of emissions were over the limits.

# 2.4. The results of dispersion calculations using climatological pollutant dispersion model

For hazards from directed and undirected sources, which concentrations exceed the allowable values maps of dispersion were drawn, considering the type

of pollutant, ground conditions, the average air temperature, thermal inversions at the moment of measurements, sizing zone and permissible limit of the pollutant in  $g/m^3$ .

The coordinate system was chosen so as to be within the entire affected area and the possible emission sources. With the help of the program used we made maps, diagrams of ground-level concentrations of pollutants, which presented the target, possibly affected neighborhoods and curves of iso-concentration for emitted pollutants. [18]

The results of dispersion calculations or maximum concentrations of pollutants at ground level (including distance from the source/site boundary) compared with the alert and intervention thresholds required by law are listed in Tables 1 and 2.

Allowed concentrations for different types of pollutants

Table 1

Pollutant	Maximu	Maximum concentration				
	$C_{max}$	Alert	The limit			
	μg/mc	threshold,	value=intervention			
		μg/mc	threshold µg/mc			
Total dust	215	75	50			
$SO_2$	260	500	350			
Total dust	170	75	50			
Total dust	109	60	40			
SO <sub>2</sub>	9	-	20			

Table 2
Concentrations of pollutants at various distances from the emission source

Concentrations of ponutants at various distances from the emission source							
Distance from the source/limit of perimeter of plaform and section (m-sector)	Concentration/span of concentrations, µg/mc	Limit value+intervention health threshold µg/mc	Observations				
	Total dust	Total dust					
0-200	215-75	50					
200-400	75-25		HOURLY				
400-600	25-5						
0-200	170-65	50					
200-400	65-15		DAILY				
400-600	15-2						
0-200	109-60	50					
200-400	60-11		YEARLY				
400-600	11-1						

# 2.5. Pollutant dispersion calculation using the mathematical model BOSANQUET PEARSON

As it was shown previously, at HELIOS Astileu the air pollution produced mainly consists of total dust resulting from production processes and particulate matter from combustion gases resulting from thermal aggregates [18].

Maximum concentration values for each source separately conducted calculation to which concentration exceeds allowable emission concentration at distances at which they were recorded, according to release calculations, are presented in Tables 3 and 4.

Values of maximum allowed concentrations of pollution

Table 3

Source	Name of	pollutant	Atmospheric	Maximum	Dist. for max.	Extreme
	pollutant		state Wind	concentration	concentration	values
			speed 1 m/s	mg/mc	m	
P1	Chimney	Total	Unstable	0.78662	150	
	cyclone	dust	Neutral	1.23028	350	0.05
	grinding		Stable	2.13172	900	
	clay					
P2	Chimney	Total	Unstable	0.14143	250	
	cyclone	dust	Neutral	0.22166	500	0.05
	trimming		Stable	0.39577	1500	
	the old ward					
P3	Chimney	Total	Unstable	0.16839	300	
	cyclone	dust	Neutral	0.26633	600	0.05
	trimming		Stable	0.48094	1500	
	new					
	department					
<b>S</b> 1	Clay dryer	$SO_2$	Unstable	0.17632	150	
	chimney		Neutral	0.27128	250	0.35
			Stable	0.46194	700	
		CO	Unstable	0.26757	150	
			Neutral	0.41168	250	10
			Stable	0.70101	700	
		Total	Unstable	0.14319	150	
		dust	Neutral	0.22030	250	0.05
			Stable	0.37513	700	
S2	Chimney of	$SO_2$	Unstable	0.09846	300	
	rotating		Neutral	0.50412	700	0.35
	furnace		Stable	0.92604	2000	
		CO	Unstable	0.07973	350	
			Neutral	0.12709	700	10
			Stable	0.23345	2000	
		Total	Unstable	0.08533	350	
		dust	Neutral	0.13600	700	0.05
			Stable	0.24983	2000	

S3	Chimney of tunnel	$SO_2$	Unstable Neutral	0.09079 0.47466	350 800	0.35
	furnace		Stable	0.87709	2000	0.55
	CT1	CO	Unstable	0.11955	600	
			Neutral	0.19182	800	10
			Stable	0.34651	2500	

 $Table \ 4$  Values of concentrations of pollutants for different sources of emission

Source	Name of pollutant	Pollutant	Atmospheric status Wind speed=3.5 m/s	Conc. Max. mg/mc	Distance for max. concentration m	Limit values
N0	Section firebrick production Share+/- 0.00	Total dust	Unstable Neutral	6.48609 8.95967	20 40	0.05
N1	Grinding department Share granulation	Total dust	Unstable Neutral	2.37587 3.28194	20 40	0.05
N2	Grinding department Share vibrating sieve	Total dust	Unstable Neutral	0.25885 0.38689	80 200	0.05
N3	Section trimming Share silos	Total dust	Unstable Neutral	0.05694 0.08690	150 250	0.05
N4	Section insulation Share metering dose	Total dust	Unstable Neutral	0.48549 0.72214	60 100	0.05
N5	Section insulation Share mixers	Total dust	Unstable Neutral	0.94734 1.36262	20 60	0.05

Dispersion calculations were made for wind speed of 0-1 m/s, which is the worst in terms of pollutant dispersion (calm air) and wind speed of 3.5 m/s (the speed that characterized that period investigated and, according to statistics, is the most frequent in the studied area.) For the given situation in which we took into consideration a slow speed of the wind (<1m/s) the calculations of dispersion were made for the three characteristics atmospheric states, (unstable, neutral, and stable) and for the speed of 3.5 m/s the calculations of dispersion were made for the three characteristic atmospheric states (unstable, neutral, stable). Dispersion

calculations were made for the pollutants with emissiond were the highest (over the allowed limits). [6]

#### 3. Conclusions

From the comparison of the results obtained from the dispersion calculations fot total dust, with maximum allowed concentration according the current legislation, we find that:

- For P1 directed source, from the clay grinding section, there were recorded the highest overruns (cca.40 times the value limit of 0.05 mg/m allowed under Order 592/2002) under conditions of atmospheric stability, and about 24.6 times in neutral atmosphere conditions.
- For directed sources P1-P2 and S12-S2 it was observed overruns between 1.6 and 14 times.
- For unguided dust emission sources we recorded overruns especially in the firebrick department and in the birebricl grinding section.
- For toxic gas resulting from combustion (S0<sub>2</sub>, CO) following dispersion calculations for directed sources, denoted by S1, S2 and S3 there resulted the following conclusions:
- For S0<sub>2</sub> it was observed exceedings of the permissible limit of 0.35 mg/cm especially in conditions of atmospheric stability.
- For CO we did not find exceedings of the permissible limits of 10 mg/m<sup>3</sup>.

Overall dedusting problem is presented as a necessity, firstly in terms of social harmfulness of dust emission, but also as a measure of economic growth in order to recover materials and rise technological operations efficiency.

### REFERENCES

- [1] Forago, I., Georgiev, K. and Havasi, A., "Advances in Air Pollution Modeling for Environmental Security" (NATO Workshop). Springer, 2005. ISSN 0957-4352.;
- [2] John Edward Smith, "Harmonization within atmospheric dispersion modelling for regulatory purposes" (8th Workshop). International Journal of Environment and Pollution, vol. 20 no. 1–6, Inderscience Enterprises, 2003. ISSN 0957-4352.
- [3] Buzduga Radu Vasile, Teza de doctorat" Cercetari privind obținerea de noi materiale refractare cu funcții complexe, realizate prin tehnologii moderne și utilizate în industria oțelului", Universitatea POLITEHNICA din Bucuresti, 2018
- [4] *Heput T, Socalici A, Constantin N., ş.a.*, "Valorificarea deseurilor feroase marunte si pulverulente", Editura PolitehnicaTimisoara, 2011, pag. 339, ISBN 978-606-554-245-7
- [5] *Ilie Butnariu, Nicolae Constantin*, "Protecția mediului înconjurător și microclimat", Editura Universitatea "Politehnica" București, 1994,137 pagini
- [6] Butnariu Ilie; Constantin Nicolae; Dobrescu Cristian; et al., "Research on the recycling of pulverulent waste from the ferous and non-ferrous industry in order to reduce the pollution", Revista de chimie, Volume: 69 Issue: 5 Pages: 1066-1070 Published: May 2018

- [7] Heput Teodor, Socalici Ana, Ardelean Erika, Ardelean Marius, "Environment ecological process in Hunedoara area through reinsertion in economic circuit of scrap and pulverous waste", Annals of the faculty of engineering Hunedoara Journal of Engineering. Tome VII (year 2009). Fascicule 3 (ISSN 1584 2673)
- [8] Socalici, Ana Virgina; Ardelean, Erika; Strugariu, Maria Laura, "Research on sustainable use of powdery waste", Environmental engineering and management journal, Volume: 15 Issue: 1 Pages: 207-212 Published: Jan. 2016
- [9] Ardelean, E.; Ardelean, M.; Heput, T.; et al., "Possibilities of recycling the lime-dolomite plant dust, Environmental engineering and management journal Volume: 11 Issue: 1 Pages: 217-226 Published: 2010
- 8] *Vonica Denisa, Mircea Hritac, Valeriu Rucai And Nicolae Constantin*; "Proposal for improvement of current technology of processing oxide ashes", in: ANNALS of Faculty of Engineering Hunedoara –International Journal of Engineering (online ISSN 1584 2673), Tome XVI (year 2018), Fascicule 3/2018 [August],BDI, http://annals.fih.upt.ro/ANNALS-2018-3.html.
- [9] *Batchvarova*, *E.*," Harmonization within atmospheric dispersion modelling for regulatory purposes" (8th Workshop), International Journal of Environment and Pollution, vol. 20, no. 1–6, Inderscience Enterprises, ISSN 0957-4352, (2003): 31-39.
- [10] *Devera*, *D*. "After installation the importance of a controlled dry-out for castable Refractories", The Refractories Engineer, (2003): 52-68.
- [11] Corneliu Stănășilă, Nicolae Constantin, Octavian Stănășilă, Raluca Petrache, "Some computations regarding directly reduction of the iron ores "U.P.B. Sci. Bull., Series B, Vol. 70, No. 2, 2008, ISSN 1454-2331,
- [12] Lipsa, F.D., Ulea, E., Chiriac, I.P., "Monitoring of fungal aerosols in some educational buildings from Iasi", Romania, Environmental Engineering and Management Journal, Vol.15, No. 4, (2016): 801-807.
- [13] Corneliu Stanasila, Nicolae CONSTANTIN, Octavian Stanasila, Raluca Petrache" A new method to dry some granular materials "U.P.B. Sci. Bull., Series B, Vol. 69, No. 4, 2007, ISSN 1454-2331
- [14] Ioana, Adrian; Constantin, Nicolae; Paunescu, Lucian; Dobrescu, Cristian; Surugiu, Gheorghe; Polifroni, Massimo; "Contribution to improving the durability of the refractory lining of the steel ladles"University Politehnica Of Bucharest Scientific Bulletin Series B-, Vol. 79, nr. 1/ 2017 ISSN 1454-2331, pag 201-220, Accession Number: WOS:000405772200020
- [15] Costoiu, Mihnea; Ioana, Adrian; Semenescu, Augustin; Constantin, Nicolae; Florea, Bogdan; Rucai, Valeriu; Dobrescu, Cristian; Polifroni, Massimo; Păunescu, Lucian; "Environmental performance indicators for decision making and stakeholder interests", Environmental Engineering & Management Journal (EEMJ),15,10,,2016, ISSN: 1582-9596, Accession Number: WOS:000392223200020, FI 1,334
- [16] *I.Butnariu*, *N.Constantin*, *C. Dobrescu*, *T.Heput*, "Research on the Recycling of Pulverulent Waste from the Ferrous and Non-Ferrous Industry in Order to Reduce the Pollution", Revista de Chimie (69), nr.5, 2018, p.1066-1070, ISSN 0034-7752, WOS 0002292756700012, [ISI-Web of Science/Science Citation Index Expanded- Factor de Impact: 1,412]
- [17] O. Dontu, N. Constantin, I. Butnariu, "Research Concerning reducing the Polluation Degrees in the Sectors of Production of Metal Materials", Revista de Chimie, vol.69, nr.8, 2018,pag 2129-2137 ISSN 0034-7752, [ISI-Web of Science/Science Citation Index Expanded- Factor de Impact 2017/2018: 1.412]
- [18] *Nicoleta Gheorghe, Cristian Dobrescu* "Laboratory research on physico-chemical properties of thermal power station slags "- Revista Metalurgia, Nr 3/2012, pag.10-14, ISSN 0461-9579