The Impact of the Industrial Processing of Oil on the Rainfall Water Quality

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Rainfall is a major component of the environment and the main source of the air purification becouse of many pollutants increases who have the most varied sources: various human activities including industry and agriculture, and some household duties. Air purification by means of precipitation is achieved by numerous highly complex mechanisms. The final products of degradation of the pollutant in the air, which are generally harmless, can be reacted with each other in the presence of water, giving rise to the final compounds with a high toxicity. Thus, exhaust, mobile sources of noxious almost identical to those specific activities in the industrial processing of oil, contain lead which is the ideal catalyst for converting SO_2 to sulfuric acid in the presence of rainwater, with all the disadvantages that they create. This paper will present an experimental research oabout how rainfall water quality is influenced by the activity of the industrial processing of oil, in a chemical plant in Constanta County.

Keywords: rainfall , air ourification, toxicity, rainwater

The increased acidification of rainfall on extensive areas in the last period, with significant negative effects on ecosystems, imposed reconsidering the place occupied by sulfur pollution of the environment [1 -3]. In the industrial areas, during certain periods of time compared with climatic factors, the concentration of SO_2 in the atmosphere exceed the long-term daily permissible average limit in our country being 0.25 mg / m^3 air [4].

In the precipitation waters, sulfur is present mainly in the form of sulphate ions. By absorbing carbon dioxide in rainwater, arise adducts acids and their negative impact on the environment is prominent [5]. Acid rainfall caused by sulfuric acid, but also of the nitric acid obtained by processes of hydration of nitrogen oxides in the presence of light, have a negative influence especially on plants, causing necrotic spots on contact, which then become vulnerable to diseases attack, finally, having a reduction in harvest levels . The negative effect of acidic precipitation is felt on metal objects containing iron, from the beginning showing rust spots, which later turns into holes, damaging and fading paints for metal objects and textiles etc.

Khanna and Beese revealed in their works the presence of high contents of SO_2 in the atmosphere, some of it goes with precipitation waters in soil, forcing its acidification. Globally, the analytical data on chemistry precipitation are calculated, processed and interpreted by WMO (World Meteorological Organization) in the program *Background Air Pollution Monitoring Network* [6, 7].

Experimental part

Knowing the chemical structure of water precipitation in the area of influence of S.C. XXX from Constanta County, has emerged as an priority objective necessity, to reflect in this way if the air was polluted with specific plant contaminant at levels above the allowable limit MAC.

The chemical composition of rainwater, and the particular degree of pollution, have certain peculiarities in time and space, being directly dependent on climatic conditions.

Thus, characteristic for the monitoring period is that was installed a quite pronounced climatic variability .

The hydro-thermal regime, showed a very high amplitude variation of previous periods, which had a marked effect, particularly on the chemical structure of rainfall in the whole area of influence (table). The rains that fell, were located in special in the extreme, being quantitatively insignificant in the warm season during most years, which caused a brief wash of the atmosphere and an excessive charging with chemical compounds, which influenced the quality of precipitation, whatever their nature. In the table 1 are shown the maximum and minimum monthly rainfall from Navodari in 2011 - 2015.

There were still large and heavy rains, almost every year and frequently exceed 100mm / 24 h, causing a very strong dilution of pollutants, regardless of the type and source of generation, which led an advanced purification of the atmosphere.

This cycle of contamination for several weeks and then quick wash was repeated several times during the warm season in the last decade, this being a feature.

Water samples of precipitation were collected monthly in May - September 2011 - 2015 in 5 points namely: NAVODARI, OVIDIU, LUMINA, SACELE and CORBU.

In terms of physico-chemical, the water quality of precipitation and thus the degree of contamination with various compounds, is not standardized in our country.

To test the degree of contamination of the water precipitation, from the plurality of possible components, were selected only few, sequenced in the three categories, namely: those which relate to the natural composition of rainwater, the toxicity and a last group comprising the

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Table 1

MAXIMUM AND MINIMUM MONTHLY RAINFALL AT NAVODARI CITY- 2011-2015

Rainfall	Months					
	IV	V	VI	VII	VIII	IX
Maxime	155.7	193.6	153.6	175.7	161.5	154.5
Minimum	16.4	8.3	21.3	34.1	11.5	1.3
Days numbers with rainfall.> 1 mm	6.6	9.0	7.4	6.5	5.6	4.3

Table 2

POSSIBLE CONSTITUENTS OF RAINFALL WATERS-(DATA COMPILED FROM LITERATURE)

Nr. crt.	Parameters	Concentrations
1.	pH	4.9 – 6.8
2.	Conductivity, µS/cm	66 – 97
3.	Chloride, mg/dm^3	4 – 10
4.	Nitrates,mg/dm^3	7 – 22
5.	Sulfates, mg/dm^3	9 – 24
6.	Phosphates, mg/dm^3	0.1
7.	Hydrocarbons, mg/dm^3	0.04 – 0.15

Table 3

THE CHEMICAL STRUCTURE OF WATER PRECIPITATION IN NAVODARI

Quality Indicator	U.M.	Month of sampling				
		MAY	JUNE	JULY	AUG.	SEPT
pH		5.91-7.40	5.35-7.40	5.15-7.80	5.35-8.55	5.27-7.40
Conductivity	µS/cm	50-104	51-192	43-70	-	51.0-115.3
Acidity	MEq/l	0.40-8.00	0.50-32.75	0.45-125.50	0.27-5.62	0.20-22.8
Ammonium, NH_4	mg/dm^3	0.01-7.25	0.01-11.33	0.01-1.35	0.001-3.860	0.03-3.70
Nitrate, NO_3	mg/dm^3	0.26-8.63	0.25-10.00	0.00-7.33	0.02-8.50	0.10-14.30
Chlorides, Cl	mg/dm^3	0.35-10.10	1.60-41.50	0.68-11.87	0.31-20.10	0.45-47.0
Sulfates, SO_4	mg/dm^3	0.01-14.55	0.08-14.00	0.10-25.50	0.07-15.00	0.0-7.0
Phosphates, PO_4	mg/dm^3	0.01-0.96	0.03-0.60	0.01-0.62	0.001-0.500	0.01-3.80
Calcium, Ca	mg/dm^3	0.20-20.0	0.1-5.0	0.00-23.30	0.1-21.0	0.1-18.0
Sodium, Na	mg/dm^3	0.40-72.40	0.28-5.65	0.10-2.33	0.35-1.80	0.22-1.85
Potassium, K	mg/dm^3	0.4-1.5	0.01-1.80	0.22-2.10	0.08-1.50	0.01-1.60
Triazines	mg/dm^3	0	0	0	0	0
Cyanide, CN	mg/dm^3	0	0	0	0	0
phenols	mg/dm^3	0	0	0	0	0
Oil product	mg/dm^3	0	0	0	0	0
CCO – Mn	mg/dm^3	3.0-4.49	7.37-16.00	1.44-16.00	1.12-3.52	2.3-4.9
CCO - Cr	mg/dm^3	56-95.9	18.0-41.2	29.2-55.3	10.0-69.0	20.0-51.0

undesirable considered compounds which can become toxic:

-natural water composition include calcium , potassium and sodium cations, whose the main source is the atmospheric dust and anions of carbonate and bicarbonate, chemical compounds that have not a high degree of dangerousness on the ecosystems in nature. These ions are found in storm water with concentration within very wide limits, in the form of areas of variation compiled from standards and papers published in the literature, as shown in table 2 - possible constituents of water precipitation.

- toxic factors include those chemicals which in low concentrations have a harmful effect on the environment. In this group of substances were tested only chemical compounds, specific to the industrial processing of oil activity which are end-products of the degradation of pollutants, such as: sulphates, chlorides, phenols and cyanides .

These anions, aggregate with nitrates as end products of hydration of nitrogen oxides, responsible for the high acidity of rainfall, have a negative effect on each segment labeled from the environment.

- the group of undesirable chemical factors, included those substances which by overcoming of a certain concentrations, have a dangerous effect to ecosystems, being subject to constant changes. Among these compounds, were selected nitrogen and phosphate anions.

Results and discussions

Taken into accont about these general aspects of chemical structure evolution of water precipitation in the influence area of the industrial processing of oil activity by SC XXX in Constanta, in the following was detailed the composition on station points, namely.

NAVODARI

Data are rpresented in table 3. The field of water rainfall chemical concentrations in Navodari in May - September 2003 – 2007, indicate the following:

- almost uniform tendency of variation of acidity rainfall throughout the period monitored;
- conductivity ranges, normally quite constant, the upper limit constant exceeding 100µS/cm;
- ammonium ions were titrated in non-uniform amounts in time and at not negligible levels;
- low concentrations of calcium, sodium, potassium, nitrogen and phosphorus ions, over time;
- small amounts of sulfates, under the variation limit specified in the literature;
- cyanide, triazines, phenols undetected in storm water.

Table 4

THE CHEMICAL STRUCTURE OF WATER PRECIPITATION IN OVIDIU

Quality Indicator	U.M.	Month of sampling				
		MAY	JUNE	JULY	AUG.	SEPT
pH	-	5.22 – 7.00	5.70-8.35	5.80-7.49	5.68-8.09	5.75-8.40
Conductivity	µS/cm	30-53	26-271	35-52	32.0-50.5	8.4-30.0
Acidity	MEq/l	0.55-30.6	0.2-113.1	0.2-128.0	0.94-78.50	0.4-27.6
Ammonium, NH_4	mg/dm³	0.01-2.80	0.01-1.37	0.005-1.370	0.004-0.900	0.001-2.300
Nitrate, NO_3	mg/dm³	0.07-9.68	0.29-15.90	0.0-10.5	0.0-6.7	0.6-12.5
Chlorides, Cl	mg/dm³	0.25-6.50	2.48-24.36	1.59-7.95	0.30-8.81	0.89-15.10
Sulfates, SO_4	mg/dm³	0.34-15.30	0.0-10.14	0.00-4.35	0.07-14.10	0.03-39.6
Phosphates, PO_4	mg/dm³	0.002-0.59	0.001-0.600	0.004-0.550	0.001-2.000	0.007-1.150
Calcium, Ca	mg/dm³	0.0-120.2	0.01-8.20	0.0-8.2	0.00-6.35	0.0-6.0
Sodium, Na	mg/dm³	0.35-2.50	0.26-1.75	0.001-1.750	0.53-1.80	0.4-2.05
Potassium, K	mg/dm³	0.06-1.60	0.10-3.00	0.01-1.80	0.01-2.00	0.01-1.80
Triazines	mg/dm³	0	0	0	0	0
Cyanide, CN	mg/dm³	0	0	0	0	0
phenols	mg/dm³	0	0	0	0	0
Oil product	mg/dm³	0	0	0	0	0
CCO – Mn	mg/dm³	4.05-34.39	5.18-14.29	1.98-14.29	3.2-4.8	5.6-8.0
CCO - Cr	mg/dm³	10.0-66.0	17.0-38.4	18-41	10-48	30-72

OVIDIU

Chemical analyzes obtained are presented in table 4. Range of concentrations of the chemical composition of water rainfall in OVIDIU in May - September 2003 - 2007 highlights the following:

- large variations, almost 1.5 units from month to month, with a tendency to acidification, of water precipitation reaction ;
- rather narrow range of variation of titratable acidity;
- decreased from month to month of rainfall water conductivity;
- very small variation of ammonium throughout the period;
- low concentrations of calcium ions, sodium, potassium, nitrogen and phosphorus over time;
- very low chloride content, located to the lower limit range considered normal;
- cyanide, triazines, phenols undetected in storm water.

LUMINA

Monitoring chemical structure at this point is shown in table 5. Chemical concentration range of waters rainfall in LUMINA in May - September 2011 - 2015 indicates:

- very close monthly variations, on the order of more than 2 *p*H units;
- the highest titratable acidity, recorded in September, from the area of influence in the monitored period;
- decrease in conductivity from spring to autumn, after a grade 2 curve;
- insignificant variations in the content of ammonium ions during the entire period of monitoring;
- low concentrations of calcium ions, sodium, potassium, nitrates and phosphates;
- insignificant movements of the variability of chloride content;
- sulfur insignificant changes in the first 4 months of monitoring and in September the range of variation to record spectacular growth;
- cyanide, triazines, phenols were not detected.

SACELE

Variations in the chemical structure of water precipitation are shown in table 6. Range of concentrations of the chemical composition of water rainfall in SACELE in May - September 2011 - 2015 indicates:

- smaller variations of reaction of rainwater in the warm seasons, with acidification tendencies;

Table 5

THE CHEMICAL STRUCTURE OF WATER PRECIPITATION IN LUMINA

Quality Indicator	U.M.	Month of sampling				
		MAY	JUNE	JULY	AUG.	SEPT
pH	-	5.80-8.05	5.43-7.90	5.10-8.22	5.60-8.07	5.70-7.72
Conductivity	µS/cm	30-53	19-78	40-102	-	22.7-50.5
Acidity	MEq/l	0.0-33,1	0.20-1.35	0.3-35.6	0.77-36.8	0.4-37.1
Ammonium, NH_4	mg/dm³	0.01-7.40	0.01-0.80	0.001-1.670	0.01-1.78	0.01-0.49
Nitrate, NO_3	mg/dm³	0.50-6.41	0.26-11.70	0.00-7.80	0.3-10.5	0.03-11.5
Chlorides, Cl	mg/dm³	0.28-10.65	0.62-15.80	2.66-21.99	0.28-10.65	2.9-11.7
Sulfates, SO_4	mg/dm³	0.01-7.39	0.01-4.76	0.06-5.60	0.00-5.80	0.11-44.95
Phosphates, PO_4	mg/dm³	0.003-1.250	0.01-0.74	0.002-0.450	0.001-0.550	0.004-0.800
Calcium, Ca	mg/dm³	0.2-5.8	0.1-35.1	0.0-6.0	0.06-14.00	0.1-14.0
Sodium, Na	mg/dm³	0.6-5.7	0.43-2.00	0.00-1.63	0.40-2.70	0.4-2.10
Potassium, K	mg/dm³	0.01-1.80	0.01-1.60	0.01-2.00	0.01-1.00	0.01-1.50
Triazines	mg/dm³	0	0	0	0	0
Cyanide, CN	mg/dm³	0	0	0	0	0
phenols	mg/dm³	0	0	0	0	0
Oil product	mg/dm³	0	0	0	0	0
CCO – Mn	mg/dm³	7.07-29.96	5.98-11.38	1.60-3.13	3.10-5.26	2.3-6.3
CCO - Cr	mg/dm³	19.1-45.9	8.0-57.6	10.0-39.1	10-66	30-92

Table 6

THE CHEMICAL STRUCTURE OF WATER PRECIPITATION IN SACELE

Quality Indicator	U.M.	Month of sampling				
		MAY	JUNE	JULY	AUG.	SEPT
pH	-	5.87-7.10	5.75-7.86	5.65-8.00	5.70-8.00	5.51-8.44
Conductivity	µS/cm	40-115	26.1-89.2	40-171	-	31.6-171.7
Acidity	MEq/l	0.8-9.1	0.71-150.7	0.9-110.5	1.1-7.9	1.4-77.0
Ammonium, NH_4	mg/dm³	0.01-5.20	0.01-2.56	0.01-3.70	0.001-0.560	0.01-2.20
Nitrate, NO_3	mg/dm³	0.12-14.80	0.15-8.50	0.00-5,65	0.01-13.00	0.04-43.00
Chlorides, Cl	mg/dm³	0.50-9.75	3.00-14.53	2.66-17.70	0.24-8.45	2.20-10.52
Sulfates, SO_4	mg/dm³	0.01-5.85	0.00-3.35	0.00-3.21	0.18-34.70	0.02-6.97
Phosphates, PO_4	mg/dm³	0.01-0.64	0.01-1.10	0.01-1.00	0.01-1.35	0.00-2.05
Calcium, Ca	mg/dm³	0.1-25.6	0.1-16.0	0.00-6.83	0.0-8.0	0.0-12.0
Sodium, Na	mg/dm³	0.4-1.5	0.25-4.80	0.01-2.12	0.3-2.1	0,26-2.85
Potassium, K	mg/dm³	0.07-1.40	0.10-2.55	0.06-3.06	0.07-1.20	0.3-2.2
Triazines	mg/dm³	0	0	0	0	0
Cyanide, CN	mg/dm³	0	0	0	0	0
phenols	mg/dm³	0	0	0	0	0
Oil product	mg/dm³	0	0	0	0	0
CCO – Mn	mg/dm³	16.34-44.93	10.78-49.7	1.70-3.13	0.9-3.1	9.6-17.6
CCO - Cr	mg/dm³	20.4-58.9	15.0-37.6	10-139	18.4-118.0	23-256

Table 7

THE CHEMICAL STRUCTURE OF WATER PRECIPITATION IN CORBU

Quality Indicator	U.M.	Month of sampling				
		MAY	JUNE	JULY	AUG.	SEPT
pH		5.78-7.40	5.80-8.41	5.50-7.77	5.90-8.00	5.70-7.70
Conductivity	µS/cm	60-141	53-127	47-130	-	27.3-202.0
Acidity	MEq/l	0.1-9.8	0.3-86.2	0.5-34.7	0.9-24.7	1.7-14.25
Ammonium, NH_4	mg/dm³	0.02-6.67	0.01-3.97	0.01-1.38	0,04-1.16	0.01-2.61
Nitrate, NO_3	mg/dm³	0.34-10.58	0.18-7.60	0.0-6.0	0.1-12.0	0.35-12.50
Chlorides, Cl	mg/dm³	0.47-8.62	1.42-9.65	1.95-13.73	0.2-9.2	1.02-28.70
Sulfates, SO_4	mg/dm³	0.01-11.25	0.00-14.40	0.16-12.60	0.04-10.20	0.05-42.15
Phosphates, PO_4	mg/dm³	0.01-0.60	0.001-2.29	0.01-0.65	0.02-1.15	0.01-1.74
Calcium, Ca	mg/dm³	0.00-8.05	0.1-4.0	0.1-10.1	0.0-8.6	0.0-7.6
Sodium, Na	mg/dm³	0.04-1.90	0.05-2.70	0.01-2.06	0.45-2.10	0.4-1.90
Potassium, K	mg/dm³	0.01-2.80	0.09-2.30	0.06-3.70	0.55-7.90	0.1-4.2
Triazines	mg/dm³	0	0	0	0	0
Cyanide, CN	mg/dm³	0	0	0	0	0
phenols	mg/dm³	0	0	0	0	0
Oil product	mg/dm³	0	0	0	0	0
CCO – Mn	mg/dm³	6.33-44.93	9.1-9.85	1.46-2.60	1.85-3.80	9.0-18.8
CCO - Cr	mg/dm³	18.0-85.8	8-48	7.9-86.0	29.2-131.0	38.4-157.0

- fairly steady range variation of conductivity recorded, with very large reduction trends;
- insignificant variations in the content of ammonium ions, calcium, sodium, and chloride, for the entire test period;
- quite constant ranges for ammonium ions;
- uneven variations for ions of calcium, sodium, phosphates, nitrates and chlorides;
- accented variations of sulphate ions only in August;
- lack of cyanide, phenols and triazines throughout the period monitored.

CORBU

Chemical analysis data are presented in table 7. Areas of concentration of the chemical composition of water rainfall in Corbu in May - September 2011-2015 highlights the following issues:

- the variation of water precipitation reaction is almost constant, with an amplitude of about 2 *p*H units, except June, when it was over 2.5 units;
- upper limit of the variation in conductivity exceeds constant 100µS/cm;
- ammonium ions have a very high amplitude variation in May, then fall accentuated by 3 times;
- very low concentrations of nitrates in July;
- almost constant variation area and reduced levels of sulfates and chlorides contents, except for September when the amplitude has accented risen.

In another paper the authors have studied the impct of the industrial processing of oil on the other components of the environmental [10, 11].

Table 8

SYNTHESIS VARIATION OF QUALITY INDICATORS IN WATER RAINFALL IN THE AREA OF INFLUENCE OF S.C. XXX IN CONSTANTA COUNTY IN MAY - SEPTEMBER 2011-2015

Number.	Quality indicators	Areas of possible concentrations	Observations
1.	pH	4.9-6.8	There have been excesses: - May - Navodari, LUMINA, SACELE, Corbu; - June - Navodari, Ovidiu, Lumina, Sacele, Corbu; - July - Navodari, Ovidiu, Lumina, Sacele, Corbu; - August - Navodari, Ovidiu, Lumina, Sacele, Corbu; - September - Ovidiu, Lumina, Sacele, Corbu;
2.	Conductivity µS/cm	65-95	There have been excesses: - May - Navodari, Sacele, Corbu; - June - Navodari, Ovidiu, Sacele, Corbu; - July - Lumina, Sacele, Corbu; - September – Navodari, Sacele, Corbu;
3.	Acidity mEq/dm^3	-	Range of variation: - Acidity: 0.08 – 14.25
4.	Ammonium, NH_4 mg/dm^3	-	Range of variation: 0.001 – 11.430
5.	Nitrate, NO_3 mg/dm^3	7-22	There have been excesses: - September - Sacele
6.	chlorides, Cl mg/dm^3	4-10	There have been excesses: - May - Oarja; - June - Navodari, Ovidiu, Lumina, Sacele; - July - Navodari, Lumina, Corbu; - August - Navodari, Lumina; - September - Navodari , Lumina, Corbu;
7.	Sulfates, SO_4 mg/dm^3	9-24	There have been excesses: - August - Călineşti, Corbu; - September - Navodari, Ovidiu, Lumina, Corbu;
8.	Phosphates, PO_4 mg/dm^3	0.1	There have been excesses: - May - Navodari, Ovidiu, Lumina, Sacele, Corbu; - June - Navodari, Ovidiu, Lumina, Sacele, Corbu; - July - Navodari, Ovidiu, Lumina, Sacele, Corbu; - August - Navodari, Ovidiu, Lumina, Sacele, Corbu; - September – Navodari, Ovidiu, Lumina, Sacele, Corbu;
9.	Calcium, Ca mg/dm^3	-	Range of variation: 0.24 – 4.65
10.	Sodium, Na mg/dm^3	-	Domeniu de variaţie: 0.34 – 1.45
11.	Potassium, K mg/dm^3	-	Range of variation: 0.27 – 1.20
12.	Triazines mg/dm^3	-	Absent
13.	cyanide, CN mg/dm^3	-	Absent
14.	phenols mg/dm^3	-	Absent
15.	Oil product mg/dm^3	-	Absent
16.	CCO – Mn mg/dm^3	-	Range of variation: 4.49 – 44.93
17.	CCO – Cr mg/dm^3	-	Range of variation: 39 – 256

Conclusions

The data and comments provided above, were summarized in table 8. Synthesis variation of quality indicators in water rainfall in the area of influence of S.C. XXX in Constanta County in May - September 2011-2015 .

Looking this table, it appears that the exceptional weather conditions of this period, the chemical structure of water precipitation was not altered as a result of nuisance pollution activity specific to the industrial processing of oil by SC XXX from Constanta.

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