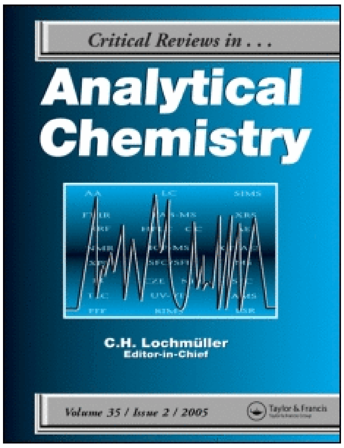

Critical Reviews in Analytical Chemistry
Publication details, including instructions for authors and subscription information:
http://www.informaworld.com/smpp/title~content=t713400837

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Online publication date: 18 June 2010

To cite this Article Misurovic, Ana(2003) 'Management of Drinking, Surface, and Underground Water Polluted by Waste from Aluminum Industry', Critical Reviews in Analytical Chemistry, 33: 4, 311 — 319

To link to this Article: DOI: 10.1080/714037683

URL: http://dx.doi.org/10.1080/714037683

Critical Reviews in Analytical Chemistry, 33(4):311–319 (2003)

ISSN: 1040-8347
DOI: 10.1080/10408340390272476

Management of Drinking, Surface, and Underground Water Polluted by Waste from Aluminum Industry

Ana Misurovic

YU Center for Ecotoxicologic Research of Montenegro, Podgorica, Montenegro

INTRODUCTION

The Parliament of the Republic of Montenegro in 1991 adopted a decision about proclaiming Montenegro to be the first Ecological State, which was also included in the Agenda 21 at the Rio Summit. The same decision was incorporated in Article One of the Montenegrin Constitution, which reads: "Montenegro is a democratic, civic, and ecological state." The rationale of such a determination by the citizens of Montenegro originated from the fact that 12% of their country's territory consists of National parks, habitats, and monuments of nature, of which two parks are listed as UNESCO's world heritage, and one is registered on the list of Ramsar Convention on Wetlands of International Importance. Montenegro's biodiversity ranks as number one in Europe in proportion with the size of its territory.

Being fully aware that the healthy drinking water is the greatest wealth of a country, and especially of a Mediterranean one (all of these coastal countries have a considerable deficit in good-quality drinking water supply), the greatest attention is being paid to the protection and management of abundant resources of so far unpolluted water.

A relatively preserved good quality of most of our surface and underground water is due to the fact that Montenegro is still industrially underdeveloped. Nevertheless, Montenegro joined many other countries in striving to achieve their industrial development, so that in the preceding period it started constructing industrial facilities by implementing inadequate technologies. The most negative example is the Aluminum Plant Podgorica (KAP).

This plant was built in the Zeta valley near the Lake of Skadar, which has been registered on the Ramsar list of protected wetland. It is the largest lake in the Balkans, and it is positioned above a unique compacted set of water springs of the highest water quality that spreads to 200 kilometers. On average, the springs feed the lake with water in the quantity of 6.34 cubic meters per sec. or 200×10^6 cubic meters per annum.

Located in the Zeta valley area is the capital town of Montenegro, as well as some 38 settlements housing a total of 140,000 inhabitants. Their activities (aluminum industry, large plantations of vineyards and orchards owned by the Agricultural company (Agrokombinat July 13), the city landfill, communal sewage collectors, and other polluters) have a considerable impact on the quality of underground water that is drained into the lake of Skadar.

The Aluminum plant operations, that is the inappropriate technologies for alumina and metal processing, as well as the improper storage of toxic waste, have the consequences of polluting a portion of the underground wells with alkaline hydroxides, fluorides, PCBs, PAHs, phenols, mineral oils, cyanides etc.

1. TECHNOLOGICAL UNITS OF THE ALUMINUM PLANT AND CAUSES OF WATER, SOIL AND AIR POLLUTION

Located in the Zeta valley, the Aluminum plant Podgorica (KAP) consists of several production units with the related service facilities: Alumina production; Anode production; Aluminum extraction, electrolysis; Foundry; Cold rolling mill; Cylumin production; Forge; Aluminum processing; Equipment maintenance unit; related facilities, such as vehicle fleet, power supply system, cafeteria, management of the plant, quality control, etc.

Alumina Processing Unit

The extraction of aluminum from bauxite is done in autoclaves by soaking them in lye (with NaOH). After the separation of the liquid content, the red mud, as gauge, is rinsed by decantation; it is then filtered, in order to extract the caustic soda. In such form, it is discharged by pipes into the waste collectors. The obtained liquid content enriched by NaOH in autoclaves, once freed from the solid content, is further processed by decomposition. Aluminum hydroxide in decomposers is, after filtering, getting calcined in order to obtain the aluminium oxide (alumina). After filtration of separated aluminum hydroxide, the liquid content is vaporized, and the concentrated solution is once again treated in autoclaves for bauxite. Upon rinsing, the red mud is disposed of in Container 1, which is coated with rubber (it did not take long for the rubber to get damaged and the container has been leaking since). Then, it is transferred to Container 2, which is uncoated. It should serve for the disposal of dry red mud with the polyelectrolyte additive for faster solidification (Giulini procedure.) In practice, the polyelectrolyte is not being added, and the mud is not dried. Namely, the moist mud (45% of moisture and 0.5% NaOH) is discharged into the uncoated container. By implementing the "Pechiney" technology, the annual production of 200,000 tons of alumina requires 500,000 tons of bauxite and 70 tons of crude oil (masut).

The main pollutants of underground waters are Aluminate, NaOH, calcined alumina, masut from the reloading ramp, and sediments from water decarbonization, which get dispersed on porous soil and enter underground waters. Caustic soda from the red mud container is drained into the ground waters.

Anode Factory

The first stage of processing is the mixing of petrol-coke and anode resin. This mixture is heated, and then pressed into raw anode blocks. A special furnace is used for the baking of those blocks in order to get baked anodes that are later used in the process of electrolysis for aluminum extraction. The anode factory has an annual capacity of 60,000 tons of baked anodes. The inputs consist of 44,000 tons of petrol-coke, 12,000 tons of anode resin and 5,000 tons of masut.

The main pollutants are dust from coke and anode residue, masut, kriolite, alumina sodium fluoride, exhaust oils, and phenol water with PAHs, which, after the rinsing of the filtering equipment for gases, are discharged into the absorption well and directly get into the underground waters.

Reduction of Aluminum—Electrolysis

Extraction of aluminum from alumina is done in electrolytic cells with the addition of NaF and kriolite as melters. Aluminum is separated on the cathode, and gasses generated in the process become tied to the anodes. The resulting liquid aluminum is vacuumed from the bottom of the cells and transferred to the foundry shop for further refinement and shaping. By the system of collectors, the gases (only in II phase) are conveyed for purification, during which fluorine is being absorbed by active alumina. Alumina, enriched with fluorine, is returned to the electrolytic cells.

The annual production of 100,000 tons of aluminum requires 44,000 tons of anodes, 2,000 tons of kriolite, and 3,000 tons of fluoride.

The main sources of pollution are permanent pollution of soil with cathode dust, kriolite, aluminum fluoride, fluorine salts, and carbonaceous matter, which are drained into the underground, and the leaking of electrolytic basins.

Aluminum Foundry

Aluminum is refined by furnacing. The refined aluminum is molded in separate equipment, in order to get desired forms. The slag extracted on the top of the furnace is collected and conveyed to the cylumin production unit. The annual production of 100,000 tons of various shapes of castings requires 1000,000 tons of liquid aluminum and 5,000 tons of masut.

The main pollutants are masut, soot, chlorine, HF, alkaline oil emulsion, slag, ammonia, and fluoride salts.

Cylumin Factory

The processing of slag and secondary aluminum to which the alloying components are added results in various aluminum alloys that are further cast into desired forms and shapes. The annual production of 15,000 tons of alloy castings requires 15,000 tons of secondary aluminum and 15,000 tons of masut.

The main sources of pollution are dump of slag and salt cakes, masut, PAHs, ammonia, fluorine salts, chlorine, HF, soot, and other aluminum waste.

Cold Rolling Mill

The cast or hot-rolled metal (Al-strips) is processed by cold rolling into strips that are used for manufacture of various products. The annual production of cold-rolled strips requires 20,000 tons of Al-strips.

The main pollutants are suspension of oils used in rolling, emulsions for grinders, and infusorial earth with oil.

Aluminum Forge

The first phase is the production of blocks for forging or pressing. The obtained blocks are placed into molds for pressing into desired forms. The annual production of 10,000 tons of forgings requires 1,000 tons of liquid Al.

The main pollutants are refiners and decarbonators, as the products of combustion, HCl, HF, MF, NaOH, graphite particles.

Aluminum Processing

This factory has several production lines. The rolling process produces a 7-micron foil or other types of thickness. Printing, polishing, pasting, etc. take place in this unit. The annual production of 10,000 of various processed items required 10,000 of Al-strips and profiles.

The main pollutants are aerosols, NaOH, rolling process oils, kerosene with additives, detergents for de-lubrication, salts of copper, zinc, and nickel.

Equipment Maintenance Unit

The pollutants are Sanitary wastewater, various waste matter, tires, transformer oil "Piralen," and Clofen (PCB-s: Arochlor 1254 and 1260), stored in barrels that leaked (9.8 tons are missing from the storage area, and 2.5 tons have leaked out), and lubricant oils that are occasionally burned (there is also a danger that "Piralen" also burned at times).

2. EXAMINATION METHODS

Examination of underground water has been going on since 1991 in over 400 household wells and piezometers in the area surrounding the Aluminum plant. Examinations have been carried out twice a year, at low and high water levels. In addition to the standard physical and chemical parameters and microbiological characteristics of water quality, special attention has been paid to the presence of PCBs, phenol, mineral oils, fluoride, cyanide, and alkaline hydroxides. Water analysis is done on unfiltered samples (except for the samples taken out from piezometers, Whatman 42), as unfiltered water from household wells is used for drinking, irrigation, and lifestock watering. All analyses are done by applying the prescribed official methods, such as EPA Methods for environmental Analysis, 1984 and EPAs Methods for Sampling and Analysis (CRC), Standard Methods for Examination of Waters and Wastewater, Edition 1989 and 19th edition, 1995. In addition to water samples, also examined are the samples of water sediments, soil, and foodstuffs, such as fish, milk, meat, eggs, cabbage "rashtan," etc. The emphasis was put on the presence of PCB-s, PCT-s content, and their congeners, as well as the presence of PCDD-s and PCDf-s, PAH-s, and fluoride, considering their ability for bioaccumulation in fatty tissue and bones (fluoride). First, analyses were done in the Agency for Health Protection in Podgorica, and after that, at the Center for Ecotoxicologic Research of Montenegro.

Phenol and cyanide analysis is done with ion-selective electrodes on apparatus ORION-701A and HORIBA F-24 and on apparatus LACHAT QC 8000 (FIA and IC), while the confirmation of results is done by spectrophotometry on apparatus Shimadzu 1601 PC VP. The analysis of heavy metals was done on apparatus Shimadzu AA-6701 PC VP with Furnace System and Shimadzu ICPS-7500 VP. The analysis of PCB-s i PCDD-s was done by gas chromatograph on instruments HP-5890 II on column HP-5 (10 m $\times$ 0.53 id mm $\times$ 2.65 μm) with ECD detector, as well as on apparatus Shimadzu GC-17AAF, Ver. 3. VP with ECD-17 detector, on column SPB-5, (30 m $\times$ 0.32 mm $\times$ 0.25 μm), while confirmation was done on Shimadzu GCMS QP-5050 Ver. 2.VP spectrometer, on column SDM-5, (30 m $\times$ 0.25 mm $\times$ 0.25 μm), u SIM-modu, along with libraries EPA, NIST, NIH, Wiley (400,000 specters). The samples are prepared by standard extraction and on SFE-400. Polycyclic aromatic hydrocarbons (PAHs) are determined by gas chromatograph on GC HP 5890 II with FID detector, on

TABLE 1
Survey of Mean Values of the Results Obtained by the Analysis of Samples Examined During 1990–2001

Samples	pH		F (mg/l)		Na (mg/l)		PCB-s (μg/l-kg)		PAH-s (μg/l-kg)		Phenols (μg/l-kg)		Mineral oils (μg/l-kg)	
	C Mid	C Max	C Mid	C Max	C Mid	C Max	C Mid	C Max	C Mid	C Max	C Mid	C Max	C Mid	C Max
Undergrou. waters														
Sector-I	7,3	7,7	0,125	1,10	4,77	17,1	1,923	20,12	1,386	9,46	0,5	2,1	23,5	386,8
II	8,24	12,31	1,44	51,56	50,41	261,9	1,98	78,10	4,23	198,1	2,9	12,6	12,11	237,6
III	7,2	7,5	0,262	1,07	5,13	13,72	0,585	2,01	2,91	11,9	1,2	2,3	0	0
IV	7,46	7,55	0,329	2,66	2,87	4,59	0,149	0,743	0,425	1,73	0,75	1.55	24,9	132,0
Surface waters														
Mora a-L							0,001	0,004	0	0				
Mora a-D							0,002	0,009	0	0				
Cijevna							0,000	0,001	0	0				
Plavnica							0,001	0,004	0	0				
Skadar lake							0,000	0,000	0	0				
Vranjina							0,001	0,004	0	0				
Canal KAP							0,000	0,002	0,32	1,78				
Sediment														
Vranjina							0,004	0,18	0	0				
Mora a-L							1,76	100,7	0	0				
Plavnica							0,12	1,25	0	0				
Skadar lake							11,76	528,4	0	0				
SOIL from							mg/kg	mg/ kg						
PiezometerB-17							0,225	381,1						
B-18							0,051	17,31						
B-19							0,125	52,28						
AS-1							10,49	245,4						
AS-2							1,23	73,50						
AS-3							14,39	144,9						
Srpska							0,23	0,469						
Botun							0,452	0,783						
Mahala							0,125	0,234						

Macrofitis from	(μg/ kg)	(μg/ kg)		
Vranjina	0,005	0,225	0	0
Mora a-L	0,038	1,71	0	0
Mora a-D	1,22	11,04	0	0
Plavnica	0,007	0,315	0	0
Skadar lake Bridge	0,025	1,125	0	0
Skadar lake Raduš	0,019	0,855	0	0
Skadar lake Grmožur	0,007	0,315	0	0
Biologic material				
Human milk	0,001	0,006	0	0
Cow milk	3,89	69,9	0	0
Eggs	207,6	451,8	0	0
Fish-Carp	2,68	713,0	0	0
Fish “jegulja”	1,3	2200	0	0
Fish “ukljeva”	5,4	498,6	0	0
Meet-cow	2,89	18,6	0,12	0,45
Cabbage	11,40	112,0	0,15	0,85

column HP-2 (10 m $\times$ 0.53 ig $\times$ 2.65 μm) and gas-mass analysis on the above mentioned instrument. The confirmation of results was done by spectrofluorometry on Shimadzu RF-1501 VP. The content of mineral oils was determined by gas chromatograph on apparatus HP-5890 II on column HP-1 along with FID detector and by FTIR analysis on Shimadzu FTIR-8701 PC VP. The conformation was done on GCMS QP-5050 in SIM Mode. All standards have been obtained from the producer SIGMA-Supelco Fluka-Aldich. Installed software programs do electronic data processing.

3. QUALITY OF SURFACE AND GROUND WATERS, SEDIMENTS, BIO-INDICATORS AND FOODSTUFFS FROM THE AREA OF ZETA

For easier presentation of results, the examined area is divided into 4 territorial units: Sector I, II, III and IV. Survey of sites where sampling was done is presented on the enclosed map. Table 1 is a survey of medium and maximum values of all measured parameters for ground and surface waters, water sediments and soil, macrophyte and foodstuffs from the endangered area.

The presented data show that the underground waters in Sector II (vicinity of the Aluminum plant—villages downstream the underground water flow) are not usable for drinking, irrigation, or animal watering, as the found values considerably exceed the permitted standards of the FRY (42/98), as well as those of the EU (80/778/EEC). Especially hazardous for human health are the extremely high concentrations of PCBs (Arochlor 1254 and 1260) that are present as ingredients in commercial products in quantities of about 3.8–20 mg/kg PCDFs (polychlorinated dibenzofurans), from 4.2 to 26 PCDDs (polychlorinated dibenzo-para-dioxins) and PCN (polyclorinated naphtalene) in concentrations of 2.6–870 mg/kg, depending on the producer of transformer oils. All mentioned substances are listed as carcinogens and mutagens with a high degree of bio-concentrations in the fatty tissue of aquatic organisms, such as fish, birds, foodstuffs, and the human body. They vary from 0.002 μg/gr in human plasma to as much as 14,000 μg/gr in the fat tissue of birds. These substances present a special danger for the ecosystem, because, as strong dielectrics, they are extremely stable. Their decomposition by chemical means or by bio-degeneration is very difficult. There are several ways by which PCBs get decomposed or dechlorinated, but none of them is applicable for underground water and the natural ecosystem. An additional risk factor for human health in this region is a high concentration of PAHs, which are also cancerous substances that break into the biological food chain. It is important to point out the fact that the solubility of PAHs (especially benzo(a)pyrene [BaP]) in water is drastically increased in the presence of phenol and organic solvents or mineral oils, which is exactly the case here, due to the water pollution from the Anode factory (the waters that after usage for the rinsing of the gasses contain both phenols and PAHs are directly drained into a drilled well). A further increase of risk to human health is that all of these toxic substances dwell in very alkaline waters with a high concentration of fluorides. Not that they are only greatly present in the ground water, but also in the air all over the Zeta valley, due to the emission from Aluminum plant, which does not have an adequate system for the capturing and filtering of fluoride gasses as a result of electrolysis.

4. PROPOSAL FOR TECHNOLOGICAL UP-GRADING

Such as endangering of environment and peril to human health in the closest neighborhood to the Aluminum plant has alerted the Montenegrin Government to form the "State Commission" with a task to define the actual risks and to propose (a) measures that would prevent further pollution, (b) changes in technologies used in some of the production units, and (c) the measures for decontamination of the endangered area. Each of the parties involved in the project were given specific assignments to that effect. The Commission defined 51 measures to be taken, out of which 31 were assigned to the Aluminum plant, which was required to implement new technologies and decontamination measures to be put in place within a five-year period. Unfortunately, the five-year term is long behind us. Meanwhile, very little, if anything, has been done in carrying out the assigned tasks.

In order to prevent the on-going migration (leaking) of alkaline waters with fluorides into the underlying strata, the initial action was to ensure permanent conservation of Container 1, which is filled up with red mud deposits from production phase one. Meanwhile, new upstanding structures have been added twice. The container should have been recultivated, or a new technology applied so that the safety of its further usage would have been ensured. Next, urgent action was required for the uncoated Container 2, where the red mud from phase two is drained. The discharge had to be immediately stopped, and the "Giulini" technology had to be fully implemented. Namely, the technology of the rinsing and filtering of red mud had to be altered to ensure the coagulation and solidification of already discharged mud by adding the prescribed polyelectrolytes, as well as the solidification of newly discharged quantities by applying the appropriate technology. Unfortunately, nothing has been done to that effect under the excuse of the lack of financing and professional expertise in these politically difficult times. Furthermore, the present condition is even worse, because the surface of the pond is constantly sprinkled with water, in order to prevent drying that would pollute the nearby settlements with red dust. The Aluminum plant has not introduced required changes in the filtering technology for gases released in either of the two phases of electrolysis, while the production has been increased and so has a degree of air pollution. No solution has been found for either the disposal of carbonated materials and coal scum or for the other anode and cathode wastes, which used to be exported and sold as a secondary raw material. The only action taken was the construction of a concrete-made covered structure for the storage of the used-up transformer oil "Piralene," so that its percolation through the ground from the damaged barrels is prevented. However, nothing was done about the required measure that all of the used up "Piralene" should be transported abroad for burning in the toxic waste incinerator or to be subjected to solidification by means of prescribed substances. The only other improvement is a new reloading ramp for masut so that it no longer gets spilled and drained into the ground water.

The rehabilitation of the polluted terrain has not even started. Furthermore, no adequate project has been developed for the clean up of soil and water contaminated by piralene oil, alkaline hydroxides, polycyclic aromatics, and other toxic substances.

A new local water supply system has been constructed in order to provide clean drinking water for the most endangered villages, but this has not brought a solution for the persisting environmental and technological problem, which also presents a challenge for scientists, because the pollution has not been stopped. On the contrary, it has been spreading in the direction of Skadar Lake.

5. PROPOSAL OF PROJECTS FOR TECHNOLOGICAL CHANGES AND REHABILITATION OF THE POLLUTED AREA

- Project for consolidation of the red mud deposited in Container 1 (the chemical composition of red mud is about 20% of aluminum trioxide, about 35% of ferric oxide, 17% of silicon dioxide, 3% of titanium dioxide, 8.5% of sodium oxide, about 3% of calcium oxide, about 45% of water content, and pH is about 12.5).
- Project for possible usage of red mud in civil engineering or for other purposes (recycling).
- Project for solidification of already deposited red mud, which is disposed of in the uncoated Container 2, in order to prevent its migration into the underlying strata. Its composition is identical to the mud in Container 1 or development of a recycling project.
- Project for prevention of further expansion of alkaline waters and other pollutants that are already present in ground waters. There is a question as to whether this can be solved by treatment with chemicals (neutralization or something similar). The hydro-technical solution is difficult or even impossible, because of the changes in the direction of the ground water courses, due to the hydrologic situation in the tributary river basis, from which the ground waters of the Zeta valley are fed.
- Project for immobilization of PCBs in soil and ground water.
- Project for treatment of smoke gases emitted by the Electrolysis plant.
- Project for treatment of carbonic substances and coal slug.
- Project for consolidation of piralene oil in storage.
- Project of recycling of all waste material from the Aluminum plant, Podgorica.

6. OTHER PROJECTS FOR CLEAN TECHNOLOGIES NEEDED BY MONTENEGRO IN ORDER TO IMPLEMENT ITS GOAL OF "MONTENEGRO—ECOLOGICAL STATE"

- Project of know-how for production of healthy food because the territory of Montenegro, with the exception of Zeta valley, is still completely unpolluted and offers great potentials.

- Production of extracts and active substances contained in medicinal herbs that abundantly grow in Montenegro, but are currently exported in only bulk.
- Production of medical drugs on an herbal basis.
- Changes in technology of steel production in the Steel Mill of Niksic.
- Technology for anaerobic digestion of household organic waste with processing into humus.
- Production of humic acid and humic fertilizers from the peat bogs in Montenegro.
- Pulp processing and the non-polluting production of paper.
- Technology for processing and cleaning of municipal and industrial waste waters.
- Projects for clean technologies applied in the secondary raw material processing (paper, rare metals, hard and soft plastics, glass, textiles, etc.)
- Technologies for safe destruction or neutralization of toxic and dangerous substances.
- Production of nonpersistent insecticides on the basis of natural products (pyrethroids and alike).
- Production of biodegradable wrappings for local ecological goods.
- Project for treatment of waste waters from wine production at the "Plantations," Podgorica.
- Project for treatment of waste waters from the edible oil plant "Primorka," Bar.
- Treatment of waste waters from daily plants in Montenegro.
- Technology for usage of rare metals, such as Ti, V, Ga, and others contained in the red mud waste.
- Technology for usage of heavy metals and rare soils from the mining waste dump of the lead-zinc and pyrite ores in Mojkovac.
- Project for usage of bauxite for production of zeolite.
- Usage of white bauxites for production of fireproof insulation and other products.

The list of possible technological and chemical treatments for making changes in currently used technologies or, for implementing the new ones, could be much longer and more specific, considering the natural resources of Montenegro.

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