

Article

Environmental Sustainability of the Alumina Industry in Western Europe

Valentina Dentoni *, Battista Grosso and Giorgio Massacci

Department of Civil and Environmental Engineering and Architecture (DICAAR), University of Cagliari, via Marengo, 2, 09123 Cagliari, Italy; E-Mails: grosso@unica.it (B.G.); massacci@unica.it (G.M.)

* Author to whom correspondence should be addressed; E-Mail: vdentoni@unica.it; Tel.: +39-070-675-5532; Fax: +39-070-675-5548.

External Editor: Marc A. Rosen

Received: 16 September 2014; in revised form: 12 November 2014 / Accepted: 11 December 2014 / Published: 18 December 2014

Abstract: The implementation of European policies on environmental protection is enforcing some substantial modifications in the processing methods and technologies traditionally adopted in the alumina industry and, in particular, in the management of the alumina residue produced. The article analyses the evolution of the alumina production and the residue disposal practices in Western Europe. Some critical aspects regarding the legal implementation of the EU Directive on the landfill of waste are highlighted and discussed. With reference to the requirements established for the landfill of non-hazardous waste, a key point is represented by the possibility of reducing the deposit protection measures if the collection and treatment of leachate is not necessary. The flexibility introduced by the Directive is not incorporated into the Italian law; this fact may represent a major issue in the prospect of disposal conversion from wet to dry methods for companies operating in Italy, as it may endanger the economic sustainability of the plants' upgrade, as well as the opportunity to attract outside investments.

Keywords: alumina industry; bauxite residue; disposal method; sustainability; European policies; Italian regulation

1. Introduction

The European policies on environmental protection are enforcing some substantial modifications in the processing methods and technologies traditionally adopted in the alumina industry and, particularly, in the management of red mud deposits. Red mud is the main residue of the bauxite treatment process for the production of alumina (Bayer process).

One of the major environmental issues for the alumina industry is the disposal of massive amounts of bauxite residue; in fact, the production of one ton of alumina leads to the generation of one to two tons of red mud [1,2], depending on the bauxite characteristics and on the processing parameters. In 2012, the global production of red mud was about 120 million tons [1–3], with an overall residue of 2.5 billion tons accumulated in land deposits. The most important environmental impact arising from red mud disposal and storage is the pollution of soil and water caused by the residue suspension fluid. The environmental risk, in fact, mainly depends on the amount of suspension fluid in the mud and on the possibility of interaction between the mud and the environmental components.

The main red mud disposal methods currently in use around the world are marine discharge, lagooning, dry stacking and dry disposal [4]. The first two methods have been in use since 1890, with the construction of the first plants for the production of alumina, whereas the adoption of the dry methods started in 1970. The general evolution of the disposal practices worldwide has shown a consistent trend away from wet to dry methods, mainly enforced by the ever-increasing strictness of the environmental protection policies. The conversion from wet to dry methods represents, in fact, a primary measure of risk containment at the source, with the reduction of the residue moisture content prior to disposal.

According to the European List of Waste [5], the red mud resulting from the alumina refining process is classified as a non-hazardous waste. The EU Directive on the landfill of waste [6] requires the disposal of non-hazardous waste in a landfill where the protection of soil and water is guaranteed by a geological barrier (natural or artificially reinforced), combined with an artificial sealing liner and a drainage layer, for the collection of leachate. The same Directive contemplates the possibility of limiting the landfill requirements to the geological barrier if the collection and treatment of leachate is not necessary or it has been established that the landfill poses no potential hazard to soil and water.

The flexibility introduced by the Directive appears quite appropriate in the general prospect of favoring the development of environmentally-sustainable activities, as it recognizes the value of the residue treatment prior to disposal and the possibility for the competent authority to reduce the provisions for the residue deposit accordingly. However, the possibilities of reducing the environmental protection measures for those companies that actually promote the environmental sustainability of their activities need to be incorporated into national law.

As a matter of fact, the Italian legal implementation of Directive 1999/31/EC [7], with reference to the landfill of non-hazardous waste, establishes the requirements for the protection of soil and water, but does not allow any simplification of the confinement barrier when the leachability is absent or negligible. The lack of flexibility in the national regulation may represent a major issue in the prospect of disposal conversion, from wet to dry methods, for the companies operating in Italy, as it may endanger the economic sustainability of the plant upgrade, as well as the opportunity to attract outside investments. In fact, other states in Western Europe (among which Germany, the U.K., France and

Spain) have incorporated into their national regulation the flexibility introduced by the European Directive. The article thoroughly discusses the issue in the frame of the current state of the alumina industry around the world and particularly in Western Europe.

2. The Alumina Industry

2.1. The Alumina Production around the World

Roughly 90% of alumina is obtained through the Bayer process, and the rest is produced by the sintering process or a combination of the two. Alumina is obtained from bauxite through the Bayer process. In the Bayer process, bauxite is heated in a concentrated solution of caustic soda (sodium hydroxide: NaOH) and sodium aluminate to temperatures between 140 °C and 150 °C for gibbsitic bauxite and between 220 °C and 270 °C for boehmitic and diasporic bauxite, under high pressure. In such conditions, a solution of sodium aluminate ($\text{NaAl}(\text{OH})_4$) is formed. After this stage (extraction), the insoluble bauxite residue is separated from the aluminum-containing liquor (green liquor) by a process known as settling. The liquor is purified as much as possible through filters before being transferred to the precipitators. The insoluble mud from the first settling stage is thickened and washed to recover the caustic soda, which is then recycled back into the main process. The green liquor is then transferred to the precipitation stage, where the crystalline aluminum trihydroxide ($\text{Al}(\text{OH})_3$) (hydrate) is precipitated. The finished mix of crystal sizes is settled from the liquor stream and separated into three size ranges in three-stage “gravity” classification tanks. The primary classifiers collect the coarse fraction, which becomes the product “hydrate”. The intermediate and fine crystals from the secondary and tertiary classifiers are washed and returned to the precipitation tanks as coarse and fine seed. The slurry of coarse “hydrate” from the primary thickeners is pumped to hydrate storage tanks and is filtered and washed on horizontal-table vacuum filters to remove and recycle process liquor, prior to calcination in fluid bed or rotary calciners. Heating the slurry before filtration, with waste steam, gives a cleaner, drier filter cake and leads to less soda loss. In the calcination phase, the “hydrate” is calcined at 1050 °C to form alumina (aluminum oxide: Al_2O_3), while water is driven off.

Bauxite is a surface alteration rock composed of a mixture of oxides and aluminum hydroxides, such as diasporite, boehmite and gibbsite, sometimes in the form of gels (derived from silicates and aluminum silicates of the original rock), with other materials of various natures, such as silica, iron oxides and hydroxides, titanium oxides, carbonates and clayey minerals. Economically, bauxite with a high content of alumina (60% to 75%), deriving from syenites and feldspathoid syenites, is the most desirable. Bauxite comes from large deposits in the sub-tropical regions of the planet (Australia, Jamaica, Guinea, South America, China, Ghana, Greece, Guyana, Hungary, India, Indonesia, Sierra Leone, Suriname, Venezuela and Vietnam). The worldwide production of bauxite is approximately 205 million tons per year (2008); ascertained reserves are 27 billion tons and estimated reserves are 38 billion [8].

Alumina production began in Europe in 1893 with the construction of plants based on the process patented by Bayer in 1892. In 1894, the first plant was built in Illinois (USA) by the Pittsburgh Reduction Company, which later became ALCOA. In 2012, the global production of alumina reached 90.17 million tons; 90% of the alumina global production is in the form of metallurgical alumina (SGA: smelter grade alumina), while the remaining 10% (NSGA: non-metallurgical grade alumina) is used for the production of zeolites, activated alumina, water treatment chemicals and several other

purposes. Bauxite processing plants are currently found in North America (11), South America (12), Europe (22), Asia (18), Australia (seven), Africa (one) and China, where the number of plants increased from seven in 2001 to 49 in 2011 [3].

2.2. Alumina Production in Western Europe

At the present time, seven plants currently in operation for the production of alumina are located in Western Europe: two in Germany (Ludwigshafen and Stade), one in France (Gardanne), one in Spain (San Ciprian), one in Ireland (Aughinish), one in Greece (Aluminum of Greece) and one in Italy (Eurallumina).

The chief characteristics of the plants are reported in Tables 1 (business name, property owner, production start-up, location and geographic coordinates) and 2 (production in 2012, number of direct employees and employees in satellite activities).

The production of alumina in Western Europe has increased since the beginning of the 1980s, with the construction of two major plants in Aughinish (Ireland) and San Ciprian (Spain); no other refineries have been built after that time. The current alumina production is about 4.11 Mt/year (2012) [9] and represents a 5% share of the worldwide production.

Table 1. The alumina plants in Western Europe.

Business Name	Property Owner	Start Up	Location	Geographic Coordinates
Ludwigshafen	Almatis GmbH	1976	Germany	49°27'6.26"N, 8°25'56.44"E
Stade	Aluminium-Oxid Stade GmbH	1973	Germany	53°38'49.59"N, 9°30'6.09"E
Gardanne	Rio Tinto Alcan	1893	France	43°27'7.92"N, 5°27'39.64"E
San Ciprian	Alcoa	1980	Spain	43°42'7.72"N, 7°28'0.69"W
Aughinish	Rusal	1983	Ireland	52°37'35.00"N, 9°3'53.00"W
Aluminum of Greece	Mytilineos Group	1965	Greece	38°21'42.56"N, 22°41'22.98"E
Eurallumina	Rusal	1973	Italy	39°11'16.04"N, 8°24'43.95"E

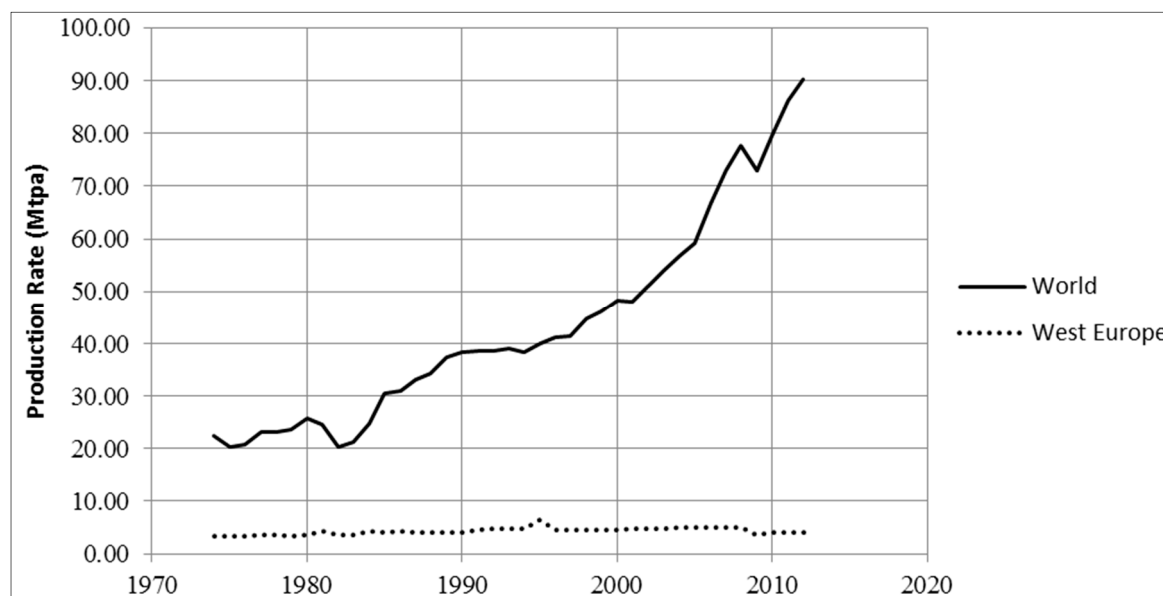
Table 2. Main characteristics of the alumina plants in Western Europe (n.d.: no data available).

Business Name	Current Production (kt/year)	No. of Employees (direct/satellite activities)	Information Source
Ludwigshafen	n.d.	n.d.	n.d.
Stade	1050	n.d.	[10]
Gardanne	460	400/250	[11]
San Ciprian	1450	1300/600 *	[12]
Aughinish	1990	450	[13]
Aluminum of Greece	800	1100	[14]
Eurallumina	0 (*)	450/250	[15]

* The production was interrupted in 2009 and will restart in 2016.

The graph of the alumina production trend in Western Europe is reported in Figure 1, where also the global production trend is represented in the same period of time (1974–2012). The comparison of the production trend in Western Europe and around the world highlights a continuous growth in global production, whereas a roughly steady level has been maintained in Western Europe.

Figure 1. World and West Europe alumina production trend (Mt/year = million tons per year).



Figures 2 and 3 represent the geographical distribution of the alumina production respectively in 1980 (for Africa, North and South America, West Europe and Oceania) and 2012 (for Africa, North and South America, West Europe and Oceania, plus China and East and Central Europe); for each productive area, both the actual output (Mt/year) and the share of the global production (%) are reported [9]. The comparison of the figures highlights a significant decrease in the Western European share of the global production, which from 14% in 1980 has become 5% in 2012, despite the growth in the production capacity (form 3.7 Mt/year in 1980 to 4.11 Mt/year in 2012), due to the relatively recent construction of the two plants in Ireland and Spain. It is worth noting that such a relevant decrease in the Western European production share mainly depends on the inclusion of the Chinese contribution, starting from 2002.

Figure 2. Geographical distribution of alumina production in 1980 (Mt/year and percentage).

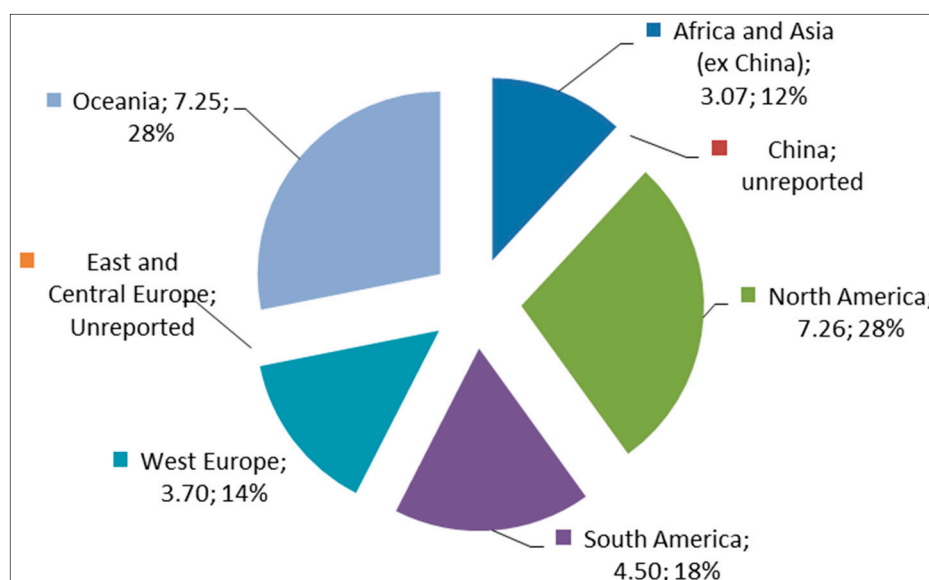
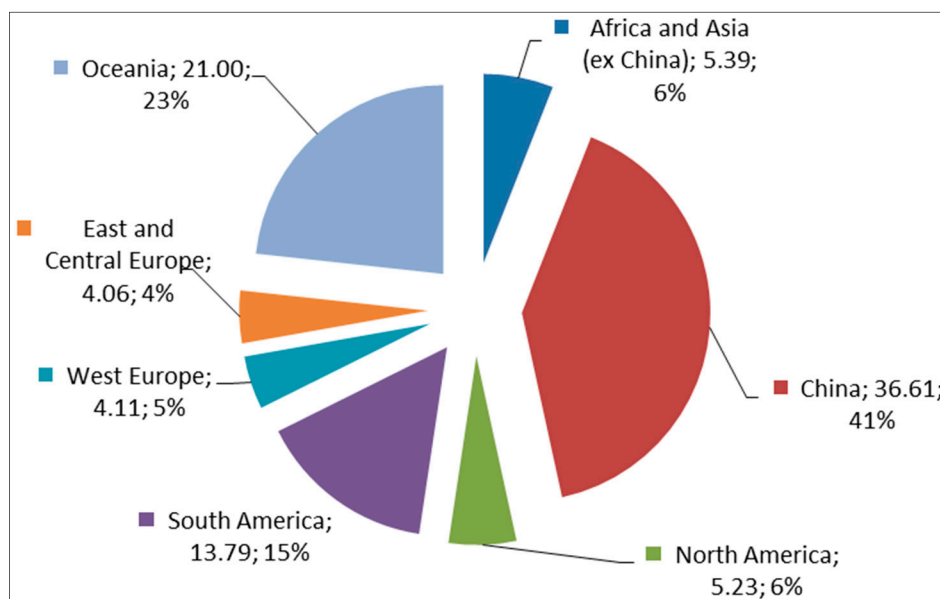


Figure 3. Geographical distribution of the alumina production in 2012 (Mt/year and percentage).

3. Bauxite Treatment and Residue Characteristics

3.1. The Bayer Process

The Bayer process for the production of alumina from bauxite includes the following phases: milling, pre-desilication (if needed), digestion, clarification and counter-current decantation (CCD) washing.

The residue of the bauxite treatment is formed during digestion and then separated from the green liquor (solution of $\text{NaAl}(\text{OH})_4$). The bauxite residue (BR) is successively transferred to the CCD washer trains, where it is repetitively washed and thickened in order to recover soda (NaOH) and alumina ($\text{NaAl}(\text{OH})_4$). After CCD washing, the BR is usually dewatered and conveyed into the bauxite residue disposal area (BRDA).

3.2. BR Characteristics

The BR chemical and physical characteristics depend on the type of bauxite used as raw material, as well as on the setting parameters in the Bayer process; therefore, different plants typically produce different types of BR.

In particular, a fundamental role in the definition of the residue characteristics is played by:

- the milling modality, as it effects the particle size distribution of the bauxite and, accordingly, its surface properties;
- the amount of desilication products (sodium alumino-silicates) introduced into the residues;
- the temperature and pressure during digestion;
- the amount of soda (NaOH) in the process;
- the type and amount of additives used during clarification to favor the separation of green liquor from the bauxite residue;
- the type and amount of flocculants used during the washing phase to recover the soda and the dissolved alumina in the remaining green liquor.

Of course, the setting of the process parameters depends on the bauxite initial characteristics. In particular, different values of temperature, pressure and soda concentration are used in the process depending on the prevalence of boehmite or gibbsite in the raw material (Table 3).

Table 3. Temperature, pressure and soda concentration for different types of bauxite.

	Gibbsite (Al(OH) ₃)	Boehmite (AlOOH)
Temperature	140–150 °C	220–240 °C
Pressure (MPa)	0.1–0.3	0.6
NaOH concentration	8.7%–3.5%	4.9%–3.5%

The BR usually contains iron oxides, aluminum oxides, titanium oxides and silica, which are originally in the raw material, plus calcium and sodium introduced during the treatment process in the form of caustic soda and burnt lime. Due to its color and consistency, the BR residue is commonly called red mud. The typical BR chemical composition is reported in Table 4, where LOI (loss on ignition) represents the organic and inorganic carbon and water that is chemically bound in the minerals.

Table 4. The typical chemical composition of the bauxite residue [16]. LOI, loss on ignition.

Red Mud Components	Fe ₂ O ₃	Al ₂ O ₃	Na ₂ O	SiO ₂	CaO	TiO ₂	LOI	Others
%	41%	16%	4%	10%	9%	9%	10%	1%

In addition to the compounds reported in Table 4, metal oxides may be present in the red mud in low concentrations or as trace elements, such as oxides of arsenic, beryllium, cadmium, chromium, copper, gallium, mercury, lead, manganese, nickel, potassium, thorium, uranium, vanadium, zinc, scandium, cerium and other rare earth element oxides.

Some of those oxides remain insoluble during the Bayer process and, therefore, can be found in the red mud; whereas soluble compounds are in the green liquor and, possibly, in the aluminum hydroxide.

Organic compounds originating from the organic matter in the bauxite can be also found in the residue and include, among others, carbohydrates, alcohols and phenols.

As mentioned before, relevant quantities of caustic soda are introduced in the Bayer process during digestion, to favor the dissolution of the aluminum compounds. The recovery of caustic soda represents a key point in the Bayer process and requires the residue to be washed in a specific section of the plant (CCD washers). The remaining caustic soda (after washing) and the soluble sodium compounds (sodium aluminate and sodium carbonate) in the liquid phase of the residue are the reason for its high pH value.

Partial neutralization by seawater lowers the pH to between 8 and 8.5 and the concentration of hydroxyl and aluminate anions, with consequent precipitation of calcium and magnesium compounds, such as calcite, aragonite, brucite, hydrotalcites, alumino-hydrocalcite, hydrocalumite and pyroaurite.

With reference to the grain size distribution, on which the physical properties of the red mud mainly depend, two classes are usually considered: the sand (grain size >100 µm) and the mud (80% of grains with a size <10 µm). The sand, if separated from the rest, is easier to wash and, therefore, assumes a low caustic content after washing; in this case, it is typically used to build internal roads within the disposal area, drainage layers and capping strata. On the other hand, if not separated from the rest, the

sand provides a better drainage capacity. The mud has a limited permeability, higher caustic content and becomes difficult to handle when the solid content is less than 70%.

3.3. Trend in the BR Treatment Prior to Disposal

With regard to the properties of the residue, it is worth noting that the best practices on the BR management give the following recommendations [1]:

- reduction and/or stabilization of the residual soda content (both in the solid residue and in the associated liquor);
- neutralization of the residue to prevent classification as hazardous material or waste;
- thickening of the residue and consequent reduction of the water content.

The cost of caustic soda represents a major component of the overall treatment cost, so that plants have always sought to recover as much caustic soda as possible for reuse within the Bayer process. There has been a progressive improvement in the recovery of the caustic soda, with a percentage of recover that is now usually greater than 96%. As for the residue solid content, recently-developed technologies, such as super thickeners, deep cone washers and vacuum drum filters, allow it to be risen from 20%–30% to about 77% [1].

The production of a ton of alumina leads to the generation of one to two tons of red mud, depending on the bauxite quality (aluminum content and type of aluminum oxide and hydroxide) and on the setting parameters during the bauxite refining process. In spite of the significant efforts spent to investigate possible options of BR reuse, the proportion of residue presently reused is negligible, and practically the total amount of BR is disposed as a waste [2].

4. EU Regulation on the Landfill of Waste

4.1. Introduction

As mentioned above, among other aspects, this article discusses the possible impact of the EU Directives on the management of the alumina treatment residue. In particular, considering that the seven European plants taken into consideration in the preceding analysis have been part of the EU since 1995, it becomes apparent that the discussion refers to the potential impact of the EU Directives on UE15 (or Western Europe).

4.2. Waste and Landfill Classification

For the purposes of Directive 1999/31/EC on the landfill of waste, the following categories of waste are defined:

- municipal waste: waste from households, as well as other waste that, because of its nature or composition, is similar to waste from the household;
- hazardous waste: any waste that is covered by Article 1(4) of the EU Directive on hazardous waste [17];
- non-hazardous waste: waste that is not covered by Article 1(4) of the EU Directive on hazardous waste [17];

- inert waste: waste that does not undergo any significant physical, chemical or biological transformations; inert waste will not dissolve, burn or otherwise physically or chemically react, biodegrade or adversely affect other matter with which it comes into contact in a way likely to give rise to environmental pollution or harm human health; the total leachability and pollutant content of the waste and the ecotoxicity of the leachate must be insignificant and, in particular, not endanger the quality of surface water and/or groundwater.

The European List of Waste (LoW) established by [5] provides a common terminology throughout the European Union with the purpose of improving the efficiency of waste management activities. The LoW serves as a common encoding of waste characteristics in a broad variety of purposes, like classification of hazardous wastes, transport of waste, installation permits, decisions about the recyclability of the waste or as a basis for waste statistics.

As regards the landfill classification, the Directive on the landfill of waste establishes that each landfill shall be classified in one of the following categories:

- landfill for hazardous waste;
- landfill for non-hazardous waste;
- landfill for inert waste.

The general requirements for each class of landfill are established in Annex I of the Directive and refer, in particular, to landfill location (Paragraph 1), water control and leachate management (Paragraph 2), protection of soil and water (Paragraph 3), gas control (Paragraph 4), nuisances and hazards (Paragraph 5), stability (Paragraph 6) and barriers (Paragraph 7).

4.3. BR Classification and Disposal According to EU Regulation

According to [5], the red mud resulting from the alumina refining process is classified as non-hazardous waste, with the identification code, 010309.

Apart from the LoW encoding, the BR physical and chemical characteristics are for the most part comparable to those of an inert waste, as defined by [6]. In fact, the red mud does not undergo any significant physical, chemical or biological transformations and does not dissolve, burn or otherwise physically or chemically react, biodegrade or adversely affect other matter with which it comes into contact. However, the pollution risk for soil and water due to the leachability and the high pH values of the bauxite residue does not allow its classification as inert waste.

As mentioned above, Annex I of Directive 1999/31/EC establishes the requirements for all classes of landfills. With reference to the protection of soil and water (Annex I, Paragraph 3), the Directive states that the landfill must be situated and designed so as to meet the necessary conditions for preventing the pollution of soil, groundwater or surface water and ensuring efficient collection of leachate. The Directive specifies that the protection of soil, groundwater and surface water is to be achieved by the combination of a geological barrier and a bottom liner, during the operational/active phase, and by the combination of a geological barrier and a top liner, during the passive phase/post closure (Annex I, Paragraph 3.1).

The Directive also establishes the possibility of completing artificially and reinforcing the natural barrier when the geological and hydrogeological conditions, below and in the vicinity of a landfill, do

not provide sufficient attenuation capacity to prevent a potential risk to soil and groundwater (Annex I, Paragraph 3.2).

In addition to the geological barrier (natural or artificially reinforced), a drainage layer and an artificial sealing liner is required for both landfill categories (hazardous and non-hazardous waste), so as to ensure that the leachate accumulation at the base of the landfill is kept to a minimum (Annex I of Directive 1999/31/EC, Paragraph 3.3).

According to the requirements of the Annex I, in absence of a natural geological barrier, the protection of soil and groundwater for a red mud basin can be guaranteed by a 1-m thick layer of clay, at the bottom of the landfill, combined with a drainage layer of 0.5 m above a High Density PolyEthylene (HDPE) geomembrane (artificial sealing liner).

Annex I, Paragraph 3.4, of Directive 1999/31/EC also states the possibility for the competent authority to reduce the landfill requirements specified in Paragraphs 3.2 (geological barrier) and 3.3 (drainage layer and sealing liner) if the collection and treatment of leachate is not necessary or it has been established that the landfill poses no potential hazard to soil, groundwater or surface water.

The flexibility introduced by Annex I, Paragraph 3.4, definitely represents the encouragement of the alumina industry towards the implementation of best available techniques (BATs) for BR treatment and disposal, as it favors the adoption of more effective methods for the recovery of caustic soda and the reduction of the residue moisture content prior to disposal. It seems quite clear, in fact, that according to EU regulation, the disposal of a residue that generates an insignificant amount of leachate and that does not endanger the quality of soil and groundwater may not require, in particular, the construction of a drainage layer and an artificial sealing liner for the collection of the leachate.

4.4. BR Classification and Disposal According to Italian Regulation

In Italy, only one alumina refinery has been operating since 1970, in the south-west of Sardinia. From that time, the residue of the bauxite treatment has been disposed by lagooning: a 25% solid residue is transferred from the refinery to the tailing basin, where the consolidation process allows the formation of a 65% solid residue.

The red mud is disposed 2 km away from the refinery in two basins. The main basin is 26 m high and occupies 114 ha of land; it has been developed according to the upstream method and presently consists of a 10 m-high lower embankment and nine secondary embankments, which give the basin its current truncated-pyramidal shape. The secondary basin is relatively recent (2005) and, therefore, composed only of the base embankment; it occupies 44 ha of land and is 10 m high.

In Italy, the legal implementation of Directive 1999/31/EC on the landfill of waste is the D. Lgs. 36/2003 [7]. With reference to the protection of soil and water, the Italian decree establishes that the deposits of hazardous and non-hazardous waste need to be provided with a geological barrier, an artificial sealing liner and a drainage layer. As regards the geological barrier, in particular, the decree states that in absence of a natural barrier or where the natural barrier is not sufficient to ensure the required attributes of thickness and permeability, an artificial confinement barrier needs to be constructed to ensure an equivalent protection for soil and water.

The decree also establishes that the characteristics of an artificial confinement barrier are normally guaranteed by coupling a layer of compacted mineral material and a geomembrane (sealing liner), but

does not contemplate the possibility for the competent authority to reduce the legal provisions depending on the actual risk for soil and water posed by the leachate. On the other hand, the term normally, which certainly refers to the common way of constructing an artificial confinement barrier, seems not to exclude different technical solutions.

Apart from different interpretations of national law, the Italian legal implementation of the EU Directive in Italy appears less flexible (the flexibility introduced by the EU Directive in Annex I, Paragraph 3.4, discussed above, is not mentioned in the Italian decree), as it institutes the use of the mineral layer-geomembrane combination for any basin of non-hazardous waste, without consideration for the presence or the actual risk posed by the leachate to soil and water.

With specific reference to the BR treatment, the significant improvement of the mud characteristics obtained through a more effective washing of the residue and by the dewatering/filtration treatment prior to disposal (practices that have been implemented by now in many plants worldwide) is not recognized by the Italian regulation. Specifically, the general trend to obtain a residue with a solid content higher than 55% (in the case of dry stacking) or 70% (in the case of dry disposal) has not generated a consequent reduction in the terms of the legal requirements for landfill, which still needs to be provided with a leachate collection system, even in absence of leachability.

The lack of a consequent reduction in the legal requirements slows the implementation of BATs for BR treatment and, at the same time, generates higher disposal costs for the alumina refineries that operate in Italy and compete with other international companies.

In fact, three out of the six countries listed in Table 1, where the alumina refineries are currently in operation, have incorporated into their national law the possibility of reducing the landfill requirements according to Annex I, Paragraph 3.4, of Directive 1999/31/EC. That is the case of Germany [18], Spain [19] and France (for the storage of mono-type hazardous waste) [20]. Apart from Italy, only Greece and Ireland have not admitted the simplification discussed above [21].

It is worth noting that in the U.K., where an alumina refinery has only recently stopped production (2002), specific statutory instruments have been adopted to guarantee the homogeneous legal implementation of the Directive's requirements (including the simplification reported in Annex I, Paragraph 3.4) all over the kingdom: England and Wales [22], Scotland [23] and Northern Ireland [24].

5. BR Production and Management in Western Europe

BR Production and Disposal

The European refineries have disposed and still dispose the BR differently, depending on the time of the production start-up, the geographical location, the residue properties and the requirements of the specific regulation regarding waste classification and disposal in the country where the plant is located. Historically, plants have disposed BR at the operating site, or in adjoining land, by filling depressions, valleys and mine workings. In some cases, when suitable sites were not available, impounding areas were created, often without a lining system, so that highly alkaline liquors reached the natural soil and groundwater. In other cases, when there were no available areas for landfill construction and depending on the distance from the coast, residues were discharged into the sea. Later, considerable improvements were introduced in the management of the BRDA, aimed at minimizing the risk of contamination and developing a closure strategy, aspects that are now a key requisite of all modern operations.

Since 1893, sea water discharge has been used in Gardanne (France) by the oldest European refinery: a 3 km-long pipeline transports the residue from the plant to the sea and discharges it at a 320-m depth into the marine Canyon de la Cassidaigne. Accordingly to the Barcelona Convention for the Protection of the Mediterranean (1975, amended in 1995), the company planned to end bauxite residue disposal into the sea by the end of 2015 and to recycle bauxite residues up to 30%, with a new product to be launched (Bauxaline®) [25]. Consequently, two filter presses have been implemented in the residue treatment process between 2007 and 2014, while a third filter press will be included in 2015 when the whole amount of produced residue will be dewatered and disposed in landfill or reused.

Since 1966, the same method has been adopted by the Viotia plant (Mytilineos Group) in the Gulf of Corinth [3]. During 2012, the group permanently dismissed the disposal of bauxite residues at sea. In the same year, the quantity of nonhazardous waste disposed in landfills was 830.000 tons [25].

Lagooning is presently operated in Stade (Germany), where, after intensive washing, the bauxite residue is pumped into pipelines to the disposal area. It is worth mentioning that, compared with other plants worldwide, the cleaning procedure of red mud operated by Aluminium Oxid Stade GmbH (AOS) results in extremely low caustic liquor losses [10]. In Italy, Eurallumina used the lagooning technique from 1977 to 2009; in fact, in 2009, the production had been suspended to integrate the dry disposal method in the production restart plan. Burntisland and Ludwigshafen represent probably the oldest European examples of dry stacking. This method has been adopted since 1941 by the British Aluminum Company in Burntisland, where residues were thickened by a plate and frame press and transported by truck to the BRDA. In Ludwigshafen, a rotary vacuum filter was adopted in 1967 to dry the residue before disposal [3]. Dry stacking has been used by other more recent plants from their start-up. In particular, in Aughinish (Ireland), the BRDA is bounded by peripheral impermeable embankments and lined, while the residue is farmed to improve water reduction. In addition, the BRDA is provided with base drains aimed at speeding mud consolidation [3]. As regards San Ciprian (Spain), the BRDA is bounded by peripheral dams, but it is not provided with lining, neither with base drains [3]. Table 5 summarizes the key information of the BR disposal for the eight plants currently operating in Western Europe.

Table 5. The main characteristics of the eight alumina plants in the EU (n.d.: no data available).

Refinery	Disposal Period	Percentage of Reuse	Disposal Method	Residue Production Rate (kt/year) *	BRDA Surface (ha)
Ludwigshafen	1976–2014	0%	Dry stacking	n.d.	6.5
Stade	1973	0%	Lagooning	1500	150
Gardanne	1893–2012	0%	Sea discharge	690	29.4
	2012–2014	30% **	Sea discharge/Dry stacking	n.d.	29.4
San Ciprian	1981–2014	0%	Dry stacking	2175	84
Aughinish	1983	0%	Dry stacking	3000	121
Mytilineos Group	1966–2012	0%	Sea discharge/ Dry stacking	1200 ***	
	2012–2014		Dry stacking	830	19
Eurallumina	1977–2009	0%	Lagooning	1200 ****	120
	From 2016	-	Dry disposal		

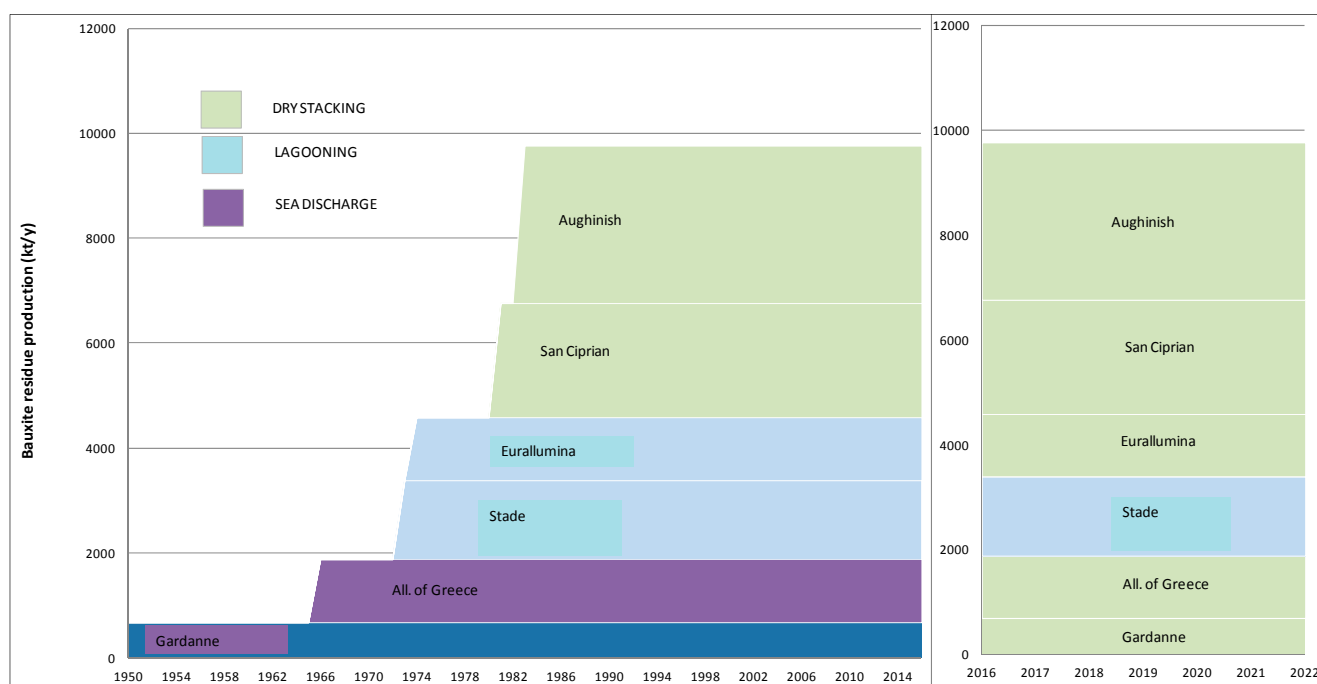
* Estimated as 1.5-times the alumina production rate [1]; ** bauxaline patent; *** in the last part of the indicated period, the amount of waste discharged into the sea was progressively reduced in favor of dry disposal; **** 2009 bauxite residue (BR) production.

As mentioned above, the potential environmental risk due to the disposal of BR is related to the physical and chemical properties of the residue, which essentially depend on the bauxite used as the raw material, on the processing parameters and on the residue treatment before disposal. In particular, the key points to be taken into consideration when considering the potential impact to soil and water are the residue water content, the water pH and the potential capacity of the solid particles to release alkaline ions into rain waters.

Because marine discharge causes direct contact between the residue fluid and the environmental component (sea water), it represents the disposal method with the highest potential impact on the environment. The potential hazard for soil and water posed by land disposal depends on the amount and quality of fluid in the mud and the presence of a geological barrier (natural or artificial) to seal the deposit and to avoid contaminant diffusion. With respect to the potential hazard posed to soil and groundwater, lagooning is the most impactful among land disposal methods.

The evolution of BR output and disposal in Western Europe is represented in Figure 4 from 1950 to 2014 (left side of the figure), together with the prospect of future conversion to dry stacking for the plants in Italy, France and Greece (right side of the figure). Figure 4 highlights that of the 10 million tons of BR produced annually from the beginning of the 1980s, 21% has been discharged into the ocean, 29% has been disposed by lagooning and 50% by dry stacking. Figure 4 also shows the improvement in the disposal practices depending on the time of the production start-up: plants set up before the 1970s (Gardanne and Viotia) adopted sea discharge; plants set up between the 1970s and the 1980s (Eurallumina and Stade) adopted lagooning; while more recent plants started with dry stacking (Aughinish and San Ciprian). It is worth noting that in Western Europe, the disposal practices have remained unchanged, even in the earliest plants, since the production start-up; this fact becomes apparent, in particular, for the French plant, where the residue was still discharged into the ocean after 120 years from its start-up.

Figure 4. Trend of BR production and disposal in Western Europe.



6. Sum-Up and Discussion

The most important environmental impact arising from BR disposal and storage is the pollution of soil and water caused by the residue suspension fluid. The environmental risk, in fact, mainly depends on the amount of suspension fluid in the mud and on the possibility of interaction between the mud and the environmental components. The most impacting disposal methods are marine discharge and lagooning. Since their start-up, the alumina refineries worldwide have been progressively adopting disposal methods with lesser impacts on the environment: from marine discharge and lagooning to dry stacking and dry disposal.

According to the European List of Waste, the red mud resulting from the alumina refining process is classified as a non-hazardous waste. The Directive 1999/31/EC requires the disposal of non-hazardous waste in landfill where the protection of soil and water is guaranteed by a geological barrier, natural or artificially reinforced, combined with an artificial sealing liner (above the geological barrier) and a drainage layer on top. The same Directive contemplates the possibility of limiting the landfill provisions to the geological barrier if the collection and treatment of leachate is not necessary or it has been established that the landfill poses no potential hazard to soil and water.

It has to be considered that, in the specific case of BR, the reduction of the moisture content to a degree that makes the residue leachability absent or negligible (*i.e.*, the collection and treatment of leachate is not necessary) may represent the possibility to classify the residue as inert waste, according to the definition of law.

After 15 years since the implementation of the EU Directive on the landfill of waste, the disposal practices adopted by the major alumina plants currently operating in Western Europe have not been significantly modified, so that by now, about 21% of the BR is discharged into the ocean and 29% is disposed by lagooning. Nonetheless, the prospect of a significant improvement in the BR disposal practices is enclosed in the long-term planning of most alumina refineries in Western Europe: by 2016, the refineries in France and Greece intend to stop marine discharge and start with the disposal of a dry residue, while the Eurallumina plant in Italy has planned to convert the current lagooning method to dry disposal [4]. As a consequence, 86% of the BR produced in Western Europe will be disposed by dry stacking or dry disposal and the remaining 14% by lagooning (Figure 4).

With reference to the landfill of non-hazardous waste, the Italian decree establishes the requirements for the protection of soil and water, but does not allow the simplification of the confinement barrier when the residue leachability is absent or negligible. As a matter of fact, the conversion from lagooning to dry disposal, included in the development plan of the only refinery currently operating in Italy, is not recognized as an improvement of the residue attributes from an environmental point of view. The company will upgrade the residue treatment process with a filtration section to drastically reduce the BR moisture content, but will not be allowed to eliminate the artificial sealing liner and the drainage layer from the basin.

In the Italian case, the general principle of favoring the development of environmentally-sustainable activities seems to be contradicted, as the strictness of the national legal requirements does not allow the reduction of the environmental protection costs in view of additional technological costs aimed at improving the residue characteristics.

7. Conclusions

The European policies on environmental protection are enforcing some substantial modifications in the processing methods and technologies traditionally adopted in the alumina industry and, particularly, in the management of the red mud deposits.

After 15 years since the implementation of the EU Directive on the landfill of waste, the disposal practices adopted by the alumina plants currently operating in Western Europe have not been significantly modified; nonetheless, the prospect of a major improvement in the BR disposal practices is enclosed in the long-term planning of most alumina refineries in Western Europe.

The key points in the implementation of EU Directives on waste and landfill of waste are the waste classification and the landfill classification. According to [5], the red mud resulting from the alumina refining process is classified as a non-hazardous waste. Directive 1999/31/EC defines the provisions for the landfill of waste in the prospect of favoring the development of environmentally-sustainable activities, as it recognizes the value of the residue treatment prior to disposal and the possibility for the competent authority to reduce the provisions for the deposit of non-hazardous waste accordingly. Accordingly, in Germany, Spain, France and the four states of the U.K., the possibility of reducing the landfill requirements according to Annex I, Paragraph 3.4, of Directive 1999/31/EC has been incorporated into national regulation.

In Italy, the general principle of favoring the development of environmentally-sustainable activities seems to be contradicted, as the strictness of the national legal requirements does not allow the competent authority to recognize a reduction in the environmental protection costs to balance the additional costs for technological innovation.

As a matter of fact, both the environmental and economic sustainability of the enterprise should to be taken into consideration by governmental authorities in charge of the approval and control of those activities that are likely to pose a threat to human health and to the environment. Evidently, the possibility of reducing the environmental protection provisions for those enterprises, which promote effective technological innovation and risk prevention at the source, needs to be incorporated into national law.

Considering the perspective of national and international industrial politics and the need for each Member State to attract outside investments, the reduction in the national regulation of the elements of flexibility contained in the EU Directives may generate a remarkable disadvantage for national enterprises, which are competing with other companies in the same field, but in different states, outside or within the EU.

Acknowledgments

The investigation was carried out in the framework of projects conducted by IGAG-CNR (Environmental Geology and Geoengineering Institute of the National Research Council), Cagliari, Italy, and by CINIGeo (National Inter-University Consortium for Georesources Engineering, Rome, Italy).

Author Contributions

Conception and design, data acquisition, analysis and interpretation and drafting of manuscript: Valentina Dentoni and Battista Grosso. Critical revision and final approval of the version to be published: Valentina Dentoni, Battista Grosso and Giorgio Massacci.

Conflicts of Interest

The authors declare no conflict of interest.

References and Notes

1. European Aluminium Association. Bauxite residue management: Best Practice. World Aluminium, April 2013. Available online: <http://www.world-aluminium.org> (accessed on 14 July 2014).
2. Klauber, C.; Gräfe, M.; Power, G. Review of Bauxite Residue “Re-use” Options. CSIRO Document DMR-3609, 2009. Available online: http://www.asiapacificpartnership.org/pdf/Aluminium/6th_aluminium_tf_meeting/Review_of_Bauxite_Residue_Re-use_Options_Aug09_sec.pdf (accessed on 14 July 2014).
3. BRaDD Bauxite Residue and Disposal Database. Available online: <https://extranet-wf.minerals.csiro.au/BRaDD> (accessed on 14 July 2014).
4. Cigagna, M.; Dentoni, V.; Grosso, B.; Massacci, G. Fugitive Dust Emissions due to the Disposal of Dried Red Mud. In *Sardinia 2013*, Proceedings of the 14th International waste management and landfill symposium, Pula, Italy, 30 September–4 October 2013; Cossu, R., He, P., Kjeldsen, P., Matsufuji, Y., Reinhart, D., Stegmann, R., Eds.; CISA: Padova, Italy, 2013; Abstract Number 332.
5. Commission Decision 2000/532/EC, OJ L 226, 06.09.2000; pp. 3–24. Available online: <http://eur-lex.europa.eu/legal-content/EN/TXT/PDF/?uri=CELEX:22002D0009&from=EN> (accessed on 14 July 2014).
6. Council Directive 1999/31/EC on the landfill of waste, OJ L 182, 16.07.1999; pp. 1–19. Available online: <http://eur-lex.europa.eu/legal-content/EN/TXT/PDF/?uri=CELEX:31999L0031&from=EN> (accessed on 14 July 2014).
7. D. Lgs. 13.01.2003 n. 36, Attuazione della direttiva 1999/31/CE relativa alle discariche di rifiuti GU 59, 12.03.2003, Suppl. Ord. n. 40; pp. 1–39. Available online: http://www.gazzettaufficiale.it/do/gazzetta/serie_generale/3/pdfPaginato?dataPubblicazioneGazzetta=20030312&numeroGazzetta=59&tipoSerie=SG&tipoSupplemento=SO&numeroSupplemento=40&numPagina=1&edizione=0&elenco30giorni= (accessed on 14 July 2014). (In Italian)
8. Power, G.; Grafe, M.; Klauber, C. Review of current Bauxite Residue Management, Disposal and Storage: Practices, Engineering and Science. CSIRO Document DMR-3608, 2009. Available online: http://www.asiapacificpartnership.org/pdf/Projects/Aluminium/Review%20of%20Current%20Bauxite%20Residue%20Management%20Disposal%20Storage_Aug09_sec.pdf (accessed on 14 July 2014).
9. World Aluminium. Available online: <http://www.world-aluminium.org> (accessed on 14 July 2014).
10. AOS Aluminiumoxid. Available online: <http://www.aos-stade.de> (accessed on 14 July 2014).
11. Alteo Gardanne. Available online: <http://www.alteo-alumina.com/en/gardanne> (accessed on 14 July 2014).
12. Alcoa San Ciprián Smelter. Available online: https://www.alcoa.com/locations/spain_san_ciprian/en/about/profile.asp (accessed on 14 July 2014).
13. Rusal Aughinish Alumina. Available online: <http://www.rusal.ru/en/about/30.aspx> (accessed on 14 July 2014).

14. AL Aluminium of Greece. Available online: <http://www.alhellas.com/en-us/home/homepage> (accessed on 14 July 2014).
15. Rusal Eurallumina. Available online: <http://www.rusal.ru/en/about/3.aspx> (accessed on 14 July 2014).
16. Gräfe, M.; Power, G.; Klauber, C. Review of Bauxite Residue Alkalinity and Associated Chemistry. CSIRO Document DMR-3610, 2009. Available online: http://www.asiapacificpartnership.org/pdf/Projects/Aluminium/Review%20of%20Bauxite%20Residue%20Alkalinity%20and%20Associated%20Chemistry_Aug09_sec.pdf (accessed on 14 July 2014).
17. Council Directive 91/689/EEC on Hazardous Waste, OJ L 377, 31.12.1991; pp. 20–27. Available online: <http://eur-lex.europa.eu/legal-content/EN/TXT/PDF/?uri=CELEX:31991L0689&from=EN> (accessed on 14 July 2014).
18. Verordnung über Deponien und Langzeitlager. Available online: <https://www.juris.de/jportal/portal/page/homerl.psml?cmsuri=%2Fjuris%2Fde%2Fkostenfreieinhalte%2Finfokostenfreieinhalte.jsp&fcstate=5&showdoccase=1&doc.part=X&doc.id=BJNR090010009#BJNR090010009BJNE003003116> (accessed on 6 November 2014). (In German)
19. Real Decreto 1481/2001, de 27 de diciembre, por el que se regula la eliminación de residuos mediante depósito en vertedero, BOE-A-2002-1697, 29.01.2002; pp. 3507–3521. Available online: <http://www.boe.es/boe/dias/2002/01/29/pdfs/A03507-03521.pdf> (accessed on 6 November 2014). (In Spanish)
20. Arrêté du 30 décembre 2002 relatif au stockage de déchets dangereux. JORF n° 90 du 16 avril 2003 page 6752 texte n° 14. Available online: http://www.legifrance.gouv.fr/affichTexte.do?jsessionid=2544EF9B6658ED2B523DA1AC655FBE27.tpdjo04v_1?cidTexte=JORFTEXT000000236168&dateTexte=20141108 (accessed on 6 November 2014). (In French)
21. Final Implementation Report for the Directive 1999/31/EC on the Landfill of Waste. 14 February 2012. Available online: <http://ec.europa.eu/environment/waste/reporting/pdf/Annex%205-1%20Landf.pdf> (accessed on 6 November 2014).
22. The Landfill (England and Wales) Regulations 2002, SI 2002/1559. Available online: http://www.legislation.gov.uk/ukxi/2002/1559/pdfs/ukxi_20021559_en.pdf (accessed on 6 November 2014).
23. The Landfill (Scotland) Regulations 2003, SSI 2003/235. Available online: http://www.legislation.gov.uk/ssi/2003/235/pdfs/ssi_20030235_en.pdf (accessed on 6 November 2014).
24. The Landfill Regulations (Northern Ireland) 2003, NISR 2003/496. Available online: http://www.legislation.gov.uk/nisr/2003/496/pdfs/nisr_20030496_en.pdf (accessed on 6 November 2014).
25. Sustainability Report 2012—MYTILINEOS Group. Available online: http://www.mytilneos.gr/Uploads/ETHSIA_DELTIA/csr_reports/Sustainability_Report_2012_en.pdf (accessed on 14 July 2014).