Chapter 29 Emission control

Two types of emissions have been blamed for creating environmental problems near aluminum smelters – fluoride and carbon compounds. These problems have ranged from crop damage and human health issues to the strength and vigor of forests and wildlife. For much of the history of aluminum production, the focus of government officials and scientists has been on fluoride. Fluoride compounds are common in the earth's crust, making up about 0.06% to 0.09% by weight. Naturally occurring fluoride is found as fluorspar, or calcium fluoride; cryolite, or sodium aluminum fluoride; and fluorapatite, or calcium fluoride phosphate. Fluorine compounds found in soils in concentrations from 20 ppm to 1,600 ppm are highly water insoluble and do not readily move through the soil. Fluorides are naturally released into the atmosphere by volcanic action, primarily in the form of hydrogen fluoride, a gas.¹

Major human sources of airborne fluoride in the U.S. have been steel manufacturers, phosphate processors, brick and tile products manufacturers, aluminum reduction plants and coal-burning operations. A 1973 journal article cited numerous incidents of fluoride emissions affecting the health of people, plant and animal life in the environment surrounding polluting plants, including the Rocky Mountain Phosphate plant in Garrison, Mont.; the Anaconda Aluminum Co. plant in Columbia Falls, Mont.; the Reynolds aluminum smelter in Troutdale, Ore.; the Ormet aluminum smelter in Hannibal, Ohio; the Alcoa and Reynolds aluminum smelters in Massena, N.Y.; the U.S. Steel plant in Provo, Utah; the Intalco aluminum smelter in Ferndale, Wash.; a phosphate plant in Polk County, Fla.; aluminum smelters in the Rhone River valley of Switzerland; the Bratislava aluminum smelter in the Soviet Union; the Kitakata aluminum smelter in Japan; and the Graz brickyard in Austria. ² This is a very short list until at least 1980, nearly every aluminum smelter in the world emitted harmful amounts of fluoride gas and particulates to its surroundings. Modern and effective air pollution systems for aluminum smelters, including use of prebake-anode type reduction pots along with primary and even secondary emissions control equipment using dry scrubbers, didn't appear until the 1970s.

The hazards of manufacturing plants that emitted fluoride to the air were known as far back as the second half of the 19th century. Early accounts of fluoride emissions damaging plant life near superphosphate plants in Europe were recorded in 1891 to 1896 and 1931; near aluminum plants in 1911 to 1918, 1934 and 1936; in chemical works in 1896, 1902 and 1931; in copper works in 1883; in iron foundries in 1931; and in brickworks in 1913. Impacts of fluorine on cattle were recorded near German zinc works in 1878; a superphosphate plant in Switzerland in 1912; an aluminum plant in Switzerland in 1911 to 1918; a superphosphate plant in France in 1928; a chemical works and a superphosphate plant in Germany in 1931; an aluminum plant in Norway in 1934; and an aluminum plant in Italy in 1935. ³ In 1968, according to the National Academy of Sciences, about 100,000 tons of fluoride were emitted to the atmosphere each year by industry around the world. That increased to about 150,000 tons per year by 1972, according to the U.S. Environmental Protection Agency. Industrial sources included manufacturers of phosphate fertilizer, phosphoric acid, elemental phosphorus, phosphate animal feed, aluminum, steel, welding, nonferrous metal foundries, brick, tile and glass factories, and by coal combustion. The largest emitters were manufacturers of aluminum, steel and brick and by coal-burning power generating plants. ⁴

European pollution problems

Prior to the discovery of the Hall-Heroult reduction process, which relies on a fluoridebased bath to dissolve alumina, aluminum was produced by a chemical process that did not utilize electrolysis but also released noxious gases. In 1855, Henri Sainte-Claire Deville joined forces with Debray, Morin and Rousseau Brothers to establish an aluminum-producing factory in Glaciere, a suburb of Paris, employing a chemical process rather than an electro-chemical process. The company soon ran into trouble with neighbors who objected to the smell of chlorine and salt fumes from the plant's manufacturing processes. ⁵ Much of the early history of fluoride emissions by other industrial plants took place in Europe. In 1855, several smelting companies in Freiberg, Germany, paid damages to neighbors injured by fluoride emissions.⁶ In 1883, scientists J.V. Schroder and O. Reuss described fluoride emissions from brick works, hydrogen fluoride plants and superphosphate plants as phytotoxicants – pollutants that harmed vegetation. H. Ost added to that information in 1907. ⁷ By 1893, the Freiberg smelting companies had paid out more than \$450,000 (in 2016 dollars) for injuries caused by air pollution and another \$400,000 for permanent relief.⁸ In 1907, emissions from the Freiberg smelters were identified as the cause of crippled cattle in the area since 1877, and fluorides were identified as the specific chemical cause. Successful lawsuits against smelting companies in both Germany and England for damages caused by fluoride emissions, along with new laws and regulations, were seen as a threat to the smelting industries.⁹ Ernest Wilczek first described crop injury near an aluminum smelter in Europe in 1916. ¹⁰

Fluoride pollution problems continued after World War I. The most notorious occurred on Dec. 3 to 5, 1930, when a deadly mist of hazardous chemicals covered the Meuse Valley of Belgium causing at least 60 deaths. "At the time of the disaster, the toxicity of fluorine compounds was little known," Kaj Roholm, a Danish scientist and the world's leading authority at the time on fluorine, reported in a 1937 journal article. "To me, however, it is quite probable that the affliction from which these people suffered was an acute intoxication by gaseous fluorine compounds emanating from certain factories in the region concerned." The 1930 Meuse Valley fog resulted from a combination of industrial air pollution and climatic conditions. The main symptom was dyspnea (shortness of breath) and the average age of those who died was 62, over a range of ages of 20 to 89 years. Cattle in the area were also affected. ¹¹ Meanwhile, studies continued of impacts to workers inside industrial plants. P.F. Moller and S.V. Gudjonsson published their findings on the physiological impacts of exposure to fluoride on Danish cryolite workers in 1932. The most severe problems were found in the vertebral column and pelvis, and the scientists noted stiffness of backs and complaints of rheumatic pains. ¹² In 1937, Roholm published "Fluorine intoxication: A clinical-hygienic study with a review of the literature and some experimental investigations." The work was referred to as a "classic treatise on fluoride intoxication" by Bertram Carnow and Shirley Conibear in 1981. Roholm described the stiffness of backs and complaints of rheumatic pains of the afflicted. ¹³

In 1889, Paul Louis-Toussaint Heroult, the French co-discoverer of the Hall-Heroult aluminum reduction process, joined with Gustave Naville, Georg Neher and Peter Huber to establish a company in Zurich, Switzerland, to produce aluminum metal. Aluminium Industrie Aktien Gesellschaft (Alusuisse) built the first aluminum smelter in Europe. The company later established plants in Neuhausen am Rheinfall, Switzerland in 1888, in Rheinfelden, Germany in 1898 and Lend, Austria in 1899. The company began to invest in the Valais valley in Switzerland in 1899 to take advantage of its hydroelectric opportunities. Pollution from the Valais aluminum plants by the 1970s created a scandal in Switzerland that went by the name "guerre du fluor" – the fluorine war. ¹⁴ Fluoride pollution from the Alusuisse smelter caused teeth and bones of livestock to become brittle, cows to produce less milk, pine trees to lose their needles, and orchards and fields to bear no yield. Fruit harvests fell from 7,000 tons to 750. The problem had been known for a long time, but by 1975 the locals had had enough and let their discontent show. The Alusuisse officials blamed bad weather. In the end, the fluorine war drew attention to both the Valais region and the problem of fluoride pollution by aluminum plants. 15

The first aluminum smelter in the Valais canton began operating in the village of Chippis in 1905. Damage to surrounding vegetation was observed by 1912. Fluoride from the Alusuisse smelters in the canton led to violence in August 1953 when apricot farmers took over a train station and set freight cars on fire. Federal troops were called in to quell the violence. That same year, another group of farmers from Fricktal brought their sick cows to the Alusuisse headquarters in Zurich, demanding compensation for damages by the company's smelter in Rheinfelden. The two groups of farmers, however, were treated differently, according to a 2013 account by Inger Weibust. Alusuisse compensated the Fricktal farmers by buying 1,580 cows over 10 years, but the Valais cantonal government was under Alusuisse influence and set extremely difficult standards for the Valais farmers. As a result, very few Valais farmers received compensation. The Valais cantonal veterinarian, forester and director of the agricultural college were believed to be influenced by Alusuisse money, according to Weibust.

Alusuisse expanded the capacity of the three Valais smelters in the 1960s, and fluoride emissions also increased. Farmers in Martigny were promised fluoride emissions would decline from 116 pounds per day to 11.4, but later testing showed the emissions actually increased six-fold. In 1968, nuns at a convent near the Chippis smelter reported headaches, dizziness and vertigo and demanded compensation. Alusuisse sent 250 Swiss francs to the Mother Superior, according to Weibust. Later, testing showed the fluoride levels in the nuns' urine was twice the level in the general population. After Alusuisse gave the convent 75,000 francs in 1975 and pledged to give them 8,000 francs per year, the nuns eventually withdrew their complaints. In 1970, after several crop failures, famers in the Martigny area founded the League Against Poisonous Factory Emissions and started sending letters to Alusuisse and authorities without result. ¹⁷

By 1975, the smelters in Valais were emitting more than 2,200 pounds of gaseous fluorine compounds per day, and the haze of pollution sometimes hung throughout the Rhone Valley. The League farmers threatened violence, and the Martigny municipal government gave the League 100,000 francs. The League then hired an agricultural expert who helped turn the group into an American-styled citizens movement, according to Weibust. When the League filed a complaint with the cantonal government, they were told the cantonal government lacked the legal authority to act and that the League would have to wait until the federal government passed an environmental protection act. Such an act did not pass until 1983. The League's agricultural expert studied aluminum smelters in Europe, North America and South Africa and then reported on the Valais smelters and the state of their pollution control measures in 1977 – providing a harsh indictment of the industry and the cantonal authorities. The League demanded hooded reduction pots and the use of dry scrubbers to deal with pot gases, and it showed that Alusuisse had lied when the company told federal authorities that its equipment retained 95% of fluoride emissions from its smelter, according to Weibust. The League's agricultural expert provided evidence showing that only 50% of the fluoride was retained.¹⁸

Alusuisse directors said it was impossible to install better pollution control equipment, claiming hooded pots and dry scrubbers were in experimental stages. Alusuisse claimed in 1976 that its plants were the most modern in Europe, but by 1976 one-third of global aluminum production took place in hooded pots, and some countries required them, according to Weibust. The League's agricultural expert also showed that Alusuisse used this kind of equipment in its own U.S. factories. Alusuisse then claimed it could not afford the 100 million francs it would cost to convert just the Chippis smelter alone. The company claimed the Chippis smelter had been operating at a loss for years, but evidence was produced showing that Alusuisse had made 427 million francs on its Swiss operations. The League's agricultural expert also cited a U.S. Department of Commerce report to say the conversion of Alusuisse's three facilities would cost only 30 million francs. Alusuisse next claimed 3,200 jobs were at risk in the town of Chippis, which had the oldest smelters, but it was later revealed that only 150 of the plant's 3,000 workers would be impacted.¹⁹

In January 1977, the Swiss federal labor agency announced that the Valais cantonal government had not been upholding a 1966 federal labor law which required that not only factory workers but the surrounding area must be protected from damaging impacts by pollution, according to Weibust. The labor agency also said the canton was responsible for enforcing the law and went on to report that the cantonal government had concealed facts from federal authorities. The cantonal government claimed it had done what it could to enforce the pollution laws, but in 1976 it had admitted it knew little about pollution control equipment in the smelters because the company never notified them about breakdowns. This admission showed the cantonal government was concerned about breakdowns, not regular operating conditions, according to Weibust. The League gave the canton a March 1977 deadline to get something done about the fluoride emissions by Alusuisse. The cantonal government announced on Dec. 12, 1977, that the Chippis plant must have permits before using new reduction pots, broken windows and leaky doors must be fixed, and the pollution control equipment must function optimally.²⁰

The League, however, was not satisfied – the requirements were considered as vague and ineffective as a cantonal law on the books since 1924. The dispute then turned violent, according to Weibust. On April 16, 1978, a transmission tower from a hydroelectric plant was blown up, farmers demonstrated and protested, and a farmer attacked a cantonal official at a meeting. A federally-appointed commission on fluoride pollution delivered a report to the cantonal executive of Valais in April 1978, but the cantonal executive kept the report secret for six months until the League discovered it. The document largely corresponded with the League's findings, reporting that the Valais smelters were barely retaining 50% of their fluoride emissions, and that it was technically possible to reduce emissions to one-quarter or one-seventh of the existing emissions. The federal report recommended reducing fluoride emissions to one-fifth of existing levels, and the Valais authorities mandated a cleanup of the three smelters in October 1978. The newer smelters at Steg and Martigny were given until 1981 to cut emissions by 80%. The three smelters at Chippis were given until 1980, 1990 and 1994 to meet the stricter standards. ²¹

In her critical history of Alusuisse, Weibust later noted that dealing with the fluoride pollution problem "was not a technical or legal problem, rather solely and entirely a political one." Writing in "Green Leviathan: The Case for a Federal Role in Environmental Policy" in 2003, Weibust observed that the Valais pollution case illustrated that local governments were not best suited to handle pollution problems. Weibust also pointed out that the negative impacts of fluoride pollution had been known since at least 1910, prevention of fluoride pollution would benefit local residents because the fluoride didn't travel far, threats of plant relocation were fairly low because the smelters needed to be near low-cost electrical power, and other aluminum plants in Europe had already made the conversion to better smelting technology so there was no cost disadvantage. "This should be an easy case for local action," Weibust said. But it wasn't.²²

The first major aluminum-producing company in England began operating on May 7, 1894, as British Aluminium Co. Ltd. (BACO). The company obtained the British and British Colonial patent rights to the Heroult process from the Societe Anonyme pour l'Industrie de l'Aluminium in Neuhausen and began aluminum smelting using hydroelectric power at Foyers, Inverness-Shire, Scotland, in June 1896 and with a second smelter plant in Argyllshire, Scotland, in December 1907.²³ By 1940, a scientist named Wilkie had published his findings on the impacts of fluoride emissions on workers at factories with fluoride emissions. He reported that two Yorkshire workmen in the production of hydrofluoric acid and aluminum fluoride were X-rayed and found to have osteosclerosis. Urinary analysis showed fluoride levels about four times what was normal. In 1945, G.F. Boddie reported on the impacts of fluoride emissions from an aluminum smelter on sheep grazing on fields within 1 1/4 miles of the smelter. Boddie found that alveolar periostitis not only made it impossible for the sheep to chew their food, but also led to infections in the skull.²⁴

In 1946, Donald Hunter published in the British Medical Journal the results of his oneyear study of the impacts of fluoride emissions from a British aluminum smelter on workers. The smelter handled 800 tons of cryolite per year, much of which was lost to the atmosphere, settling in particulate form on surrounding fields where grazing sheep and cattle developed fluorosis. Hunter also found that 28 of 264 "furnace-men" had skeletal fluorosis, although none had complained of it. In the Dec. 7, 1946, issue of the

British medical journal The Lancet, Margaret Murray and Donald C. Wilson reviewed literature on the impacts of fluorine emissions from industry on workers, residents and plants and animals. A recent outbreak of fluorosis in cattle in England "has once more drawn attention to the large amount of fluorine and fluorine compounds being set free by some recently extended industrial processes and has shown the necessity for consideration of the dangers to public health and to agricultural economy existing in the neighborhood of these undertakings." Citing studies dating back to 1937, Murray and Wilson concluded, "That fluorine must be regarded as a cumulative poison is well established by chemical and experimental observations." The two recommended using pollution control equipment to limit fluoride emissions.²⁵ Some company changes were evident four decades later. When construction of the Lynemouth aluminum smelter began in northeast England in 1974, local farmers were worried that pollution from the plant would ruin their crops and harm their livestock. To address the farmers' concerns, Alcan decided to buy the land from them. Alcan acquired more than 4,500 acres of land in the local area and even employed a farming director. The land continued to be used to grow crops and raise livestock in 2016.²⁶

In 1954, an aluminum reduction plant began operating in a remote narrow valley near Sunndalsora, Norway, operated by Ardal og Sunndal Verk. Soon after the smelter began operating, fluoride pollution from the plant caused damage to agricultural and other lands. By 1969, the industrial complex had grown to be the largest aluminum smelter in Western Europe, and local farmers had collected about \$500,000 in damages. The company used enlightened social policies to deal with most of its problems, including guarantees against layoffs, generous allowances for private housing and saunas for the workers, according to the New York Times. Located at the end of a 62-mile long fjord bounded by 5,000-foot high mountains, the air pollution problem was aggravated by the direction of prevailing winds in summer. The problem worsened when the plant doubled in size in 1968. One local farmer claimed that the lower valley was "totally destroyed for cattle and forests." But most farmers bore the company no ill will and praised the company's attempts to settle claims. The plant spent \$3.4 million on air pollution control equipment and planned to spend \$2.1 million more in 1969. Prohibitions on new reduction pot start-ups were in effect during the growing seasons.²⁷

The technological blame for much of the Norwegian air pollution was placed on Soderberg-type reduction pots. On Feb. 14, 2003, Norsk Hydro announced it would shut down up to 200,000 tons per year of aluminum smelting capacity at three of its plants in Norway that used Soderberg anodes because the plants were unable to meet environmental norms. The shutdown amounted to about one-quarter of Norsk Hydro's total capacity in Norway. By 2006, about 50,000 tons at Aardal and about 20,000 tons at Hoyanger would be shut down. Another 120,000 tons would be shut down at Karmoy by the end of 2009. The Aardal plant had a total of 200,000 tons capacity, the Hoyanger plant had 73,000 tons capacity, and the Karmoy plant had 267,000 tons capacity. The rest of the plants used a different anode technology and could meet environmental norms, the company said. Norsk Hydro at the time was the world's third largest aluminum group and the largest in Europe, with a global smelting capacity of 1.4 million tons. ²⁸ Alcoa shared Norsk Hydro's concern about Soderberg pots. On Oct. 25, 2004, Alcoa announced plans to spend \$79.4 million upgrading technology and environmental equipment at its aluminum smelters in Aviles, Coruna and San Ciprian, in Spain. The money would be used to reduce emissions of fluoride, dust and benzo(a)pyrene at the Aviles plant, which used Soderberg pots. Alcoa's alumina refinery at San Ciprian was also included in an agreement with the Galician government to improve environmental safeguards. ²⁹

Canadian fluoride cases

Fluoride emissions from two U.S. smelters in Massena, N.Y., were blamed for damage to cattle and human health after the pollution blew across the international border into Canada. Two Canadian government agencies began to have concerns over a link between ecological impacts on the St. Regis Akwesasne Indian Reserve and fluoride emissions by the Reynolds Metals Co. plant in Massena between 1969 and 1971. Mohawk Indians claimed they noticed increased livestock losses, dying crops, dying trees and even dead bees on Cornwall Island shortly after the smelter began operating. ³⁰ In 1959, about 45 farmers with 40 cattle barns and 364 dairy cattle lived on the island in the Gulf of St. Lawrence, between Ontario and New York. That year, Reynolds began operating an aluminum smelter on the south bank of the St. Lawrence River near Massena, which was upwind from Cornwall Island about 60% of the time. According to Angus Lazores, a Mohawk Indian resident, cattle on the island soon became lame and developed swelling on their legs to the point where they could no longer graze. They lay down on the pasture to eat and crawled from one place to another, he said. ³¹

In 1971, S.N. Linzon reported in the Proceedings of the Second International Clean Air Congress on fluoride effects to vegetation in the Cornwall Island area that was downwind from two large aluminum plants and a fertilizer plant. Foliar fluoride concentrations up to 396 ppm were found within one mile of one of the aluminum plants. For his study, Linzon established 35 ppm as the threshold concentration for injury to plants. All of the vegetation he studied was within 4 1/2 miles of the industrial fluoride sources, so he had no controls and he did not cite a baseline level. ³² By November 1971, most farmers on the island had switched from dairy cattle to beef cattle, but only 177 cattle remained. Officials at the Canadian Ministry of the Environment had expressed their concerns to Reynolds about fluoride emissions causing the problems as early as 1969. The St. Regis Local Council authorized an investigation into the emissions in 1973 and was advised in July 1973 that damage to pine trees on the island was caused by fluoride gases. Urine samples collected from cattle in 1975 showed abnormal levels of fluoride. ³³ Laboratory evidence of the impacts of fluoride on plant life near aluminum plants became available in 1973 when T. Facteau, S.Y. Wang and K.E. Rowe reported in the Journal of the American Society for Horticultural Sciences on laboratory experiments where plants were fumigated with hydrogen fluoride. They found that using 2 to 3 ppb fluoride in their experiments, similar to atmospheric levels near aluminum smelters, reduced pollen germination and pollen tube growth. ³⁴

In 1973, Reynolds installed \$17.8 million worth of pollution control equipment at its Massena smelter and reduced the Massena plant's fluoride emissions from more than 7,200 pounds per day to 2,688 pounds, and later to 1,776 pounds. Conditions on Cornwall Island improved but did not disappear.³⁵ The Reynolds smelter emitted about 286 pounds of fluoride per hour from 1959 to 1969, at which point New York State regulations forced the plant to reduce its emissions to 66 pounds per hour, or 1,584 pounds per day, by 1975. Reynolds also sent a veterinarian to inspect cattle on Cornwall Island in November 1975. The veterinarian claimed that internal and external parasites were responsible for the cattle's condition – fluoride was not mentioned in the veterinarian's report. Concerned about the veterinarian at Cornell University. After extensive diagnostic and pathological testing, Krook announced, "Owing to the extensive and serious chronic fluoride poisoning, no cattle born on Cornwall Island were going to live more than five years." ³⁶

The situation was further investigated by scientists at Cornell University in 1977 and 1978. In their report, Krook and George Maylin said, "Of all pollutants that affect farm animals, fluorine has caused the most severe and widespread damage." Clancy Gordon, a plant pathologist at the University of Montana, examined 2,600 plant samples from Cornwall Island and found very high levels of fluoride in all the samples. Bertram Carnow and Shirley Conibear, from the University of Illinois, examined the health of the island's residents and found "significant numbers of people with abnormalities of the muscular, skeletal, nervous and blood systems." Physicians who looked at residents earlier had reported high rates of anemia, rashes, irritability, diabetes, high blood pressure and thyroid disease. "Unquestionably, heavy exposure to fluorine compounds has affected all the life studied," Carnow and Conibear reported. ³⁷ In April 1979, findings of extensive fluoride testing by Krook and Maylin appeared in the Cornell Veterinarian journal. Injuries to the cattle were blamed on fluoride emissions, and the two concluded that the 40 ppm standard for fluoride in forage that was recommended as a safe upper limit by the National Academy of Sciences was anything but safe. Krook

and Maylin reported finding damage to cattle in New York when the fluoride levels in forage were as low as 13 ppm to 24 ppm.³⁸

A 1978 study by Carnow and Conibear commissioned by the Indian tribe concluded that Clancy Gordon's evidence of fluoride in vegetation samples showed the Reynolds smelter was responsible for the high concentrations of fluoride on the island. The Carnow-Conibear report also included a two-day survey of Cornwall Island residents which found evidence of muscular, skeletal, nervous and blood abnormalities, and school children who were irritable and hyperactive. Two years later, an epidemiological study of the impacts was begun by the Mount Sinai School of Medicine in New York City. In February 1980, the Indian tribe filed a \$150 million lawsuit against Reynolds and Alcoa, which also operated an aluminum smelter in Massena.³⁹

In April 1980, a report in the Cornell Veterinarian journal claimed New York State and federal fluoride emission standards did not protect cattle. The conclusions were based on studies of cattle on Cornwall Island where ambient airborne fluoride never exceeded New York standards on the island, and fluoride contamination in forage was well below 40 ppm, which was the tolerance level set by the National Academy of Sciences. Nevertheless, 63 of the 82 dairy cattle on the island were slaughtered in 1979 because of chronic fluoride pollution. The cattle showed increased evidence of fluorosis as they got older, including stunted growth and dental fluorosis that interfered with drinking and chewing. The Reynolds plant had emitted about 1,600 pounds of fluoride per day since 1973, and considerably much more from 1959 through 1973, the journal article reported. ⁴⁰ Cornwall Island was not the only site in Canada threatened by fluoride emissions from factories. In 1977, S.S. Sidhu reported on abnormally high fluoride levels in air, vegetation and soil near a phosphorus plant in Newfoundland. He concluded that atmospheric fluoride levels from 0.20 to 0.25 ppb no more than 60% of the time appeared to be "safe" levels for forest species. His report was presented at the 70th Air Pollution Control Association meeting in Toronto, Ontario.⁴¹

Communist fluoride emissions

In the former Soviet Union, whole regions suffering from pollution and ecological collapse resulted from decades of badly planned industrial and military development that were undertaken in virtual secrecy and with little concern for environmental and health consequences. Pollution in Russia following the collapse of the Communist government threatened the health of millions of residents and the safety of crops, water and air. Air pollution was 10 times accepted safety levels in 84 of Russia's largest cities. In some areas, especially among children, levels of respiratory problems were 50% higher than the national average. ⁴² The nation's huge aluminum industry was to blame from some of this pollution. In October 1993, the Bratsk smelter in Irkutsk,

Siberia, in Russia, provided the livelihood for about 300,000 city residents with 20,000 direct jobs and 77% of the city's income. The smelter used outmoded Soderberg anode technology and was considered a major polluter. At the time, the plant was producing 750,000 tons of aluminum per year, well below its capacity of 840,000 tons.⁴³

In December 1993, Bratsk officials announced the aluminum plant would become an ecologically safe facility by 2000 by upgrading reduction pots and installing air pollution control equipment. Evidence of air pollution impacting trees 12 miles to the west and up to 18 miles to the east had been reported in 1985. The Bratsk Forestry Association had filed claims against the smelter with the USSR State Board of Arbitration, which levied a 21 million ruble fine with no result. The plant had paid large fines in the past. In June 2001, Russian NTV broadcast a report on the Bratsk smelter and attempts to move residents to a new location. Chikanovskiy, a village not far from the smelter's smokestacks, was described as a "ghost village" after 40 years of smelting. A district health officer said residents experienced changes in the calcium content of their bones and suffered from bronchitis. The Russian TV correspondent said the residents should have been relocated in 1975 "but the process began only in 1994" when 643 families were moved. The relocation program was to be financed by the government and the plant, but the smelter claimed it had no money.⁴⁴

Soviet scientists had been well aware of the problems caused by aluminum plant emissions. From 1973 through 1987, Russian scientists A.S. Rozhkov and T.A. Mikhailova studied the effects of fluoride emissions on conifers at the Laboratory of Pathology of Woody Plants at the Siberian Institute of Plant Physiology and Biochemistry in Irkutsk. Siberian larch trees ranging from 10 to 12 years old were placed in chambers and exposed to hydrogen fluoride, chlorine, sulfur dioxide, carbon monoxide, nitrogen oxides, or combinations of these gases for 18 hours per day. Fluoride-containing emissions proved to be the most dangerous to the plants. "Fluorine derivatives are the most aggressive among toxic compounds polluting the atmosphere," they said in their 1993 book on the subject. "Moreover, the percentage of fluorides in industrial emissions is constantly increasing, with the bulk of the fluorides being emitted by aluminum smelters. Fluorine is poorly detoxified by both plants and animals, and the accumulation of even relatively low concentrations over a long period causes a cumulative toxic effect. Among woody plants, conifers are less resistant to fluorine." ⁴⁵

The impacts caused by the Soviet Union's industrial and military facilities extended beyond the Russian borders. On Nov. 17, 1994, the governments of the Republic of Uzbekistan and the Republic of Tajikistan signed an agreement to cooperate in improving the environmental conditions in the area surrounding the Tajik Aluminum Plant. According to Karshi Sulanov, a district health officer commenting in 2000, nothing came out of the agreement. "The frightening thing is that Tajikistan, while fully aware of the aluminum plant's impact on the environment and public health, isn't taking any steps whatsoever to prevent these negative effects," Sulanov said. The Tajik Aluminum Plant was one of the former Soviet Union's largest aluminum smelters, producing up to 514,000 tons per year while consuming about 40% of Tajikistan's electrical power and emitting about 193 tons of fluoride, 1,306 tons of sulfur dioxide and 28,900 tons of carbon monoxide per year. In 1991, the Russian Press Digest reported that the aluminum plant's surroundings were "environmental disaster areas." Cows had missing teeth, silkworm production had stopped, and vineyard, pomegranate and stone fruit crops had all perished. In July 1995, the BBC reported that the Tajik Aluminum Plant had purchased \$4.5 million worth of air pollution control equipment from a Norwegian company. By 2000, a shortage of raw materials had caused the plant to reduce production to about 38% of capacity. ⁴⁶

Eastern Europe also experienced a significant amount of environmental damage in the 20th century as the result of war and totalitarian governments. In 1953, Jan Lezovic began studying the impacts of fluoride emissions from an aluminum smelter in Bratislava, Czechoslovakia, on surrounding vegetation in the three-mile wide mountain valley where the smelter operated. The nearest village was about 300 yards away. Smoke, vapors and solid particles contaminated the atmosphere, and the 1966 harvest was less than one-half the yield of the year prior to operation of the smelter. Leaves on fruit trees and garden vegetables were harder, glossier and tougher, yet more fragile than normal and covered with a whitish-gray crust. Fluoride was 20.9 times normal in apples and 10 times normal in lettuce. Fluorine levels in snow deposits were significantly high. Two years after the plant began operating, 95% of cattle and goats in the valley were afflicted with fluorosis, and all 70 bee colonies died off. ⁴⁷ On Jan. 3, 1981, the mayor of Cracow, Poland, ordered the closure of half the Skawina aluminum smelter's capacity on environmental grounds. According to the monthly newsletter Prawo i Zycie (Law and Life), the smelter emitted 92.4 pounds of fluoride per ton of aluminum produced in 1980, when most Western smelters did not emit more than 3.3 pounds per ton. There was a particularly high incidence of inflammation of respiratory passages in people near the smelter, and a court action was brought by nearby residents in November 1980. The Solidarity government decided the cost of modernizing the smelter was too high. ⁴⁸ The smelter, which began operating in 1954 as Huta Aluminium Skawina southwest of Karako, was shut down for environmental reasons in 1981.

The rapid growth of the Chinese economy since the start of the 21st century has impacted the country's air and water, especially as more and more factories have relied on coal-fired generators and unenforced regulations have led to abuse by factory managers. Growth in the aluminum industry has been extraordinary. China's share in

consumption of primary aluminum increased from 2% of the global market in 1972 to 40% in 2012. ⁴⁹ Production followed suit, with China producing 23.3 million tons of the world's 49.3 million tons by 2014.⁵⁰ Emissions from China's aluminum smelters have impacted far-flung rural areas. In February 2001, reports were made about fluoride pollution in eastern Tibet from the Qinghai aluminum smelter near the Chinese city of Xining, which was owned by the Chinese Aluminum Co., Chalco. In February 2001, Gabriel Lafitte reported in The Age about Tibetan farmers complaining of "smoke" from the smelter settling on hillsides and causing the teeth of grazing donkeys and sheep to turn yellow and become brittle. Kaiser had joined Chalco in the aluminum venture but pulled out later because of insufficient hydroelectric power. In December 2001, Students For A Free Tibet sent a letter to the U.S. investment banker Morgan Stanley protesting the sale of stocks for Chalco. The group pointed out that an "independent Australian engineering organization" had found that the Xining smelter exceeded government standards for fluoride emissions, and the group described sheep in nearby Tibetan villages losing their teeth after grazing on fluoride-contaminated grass. The Central Tibetan Administration also wrote to Morgan Stanley about the Xining smelter. The letter said Worley Chemicals & Minerals conducted an environmental review and found that the Xining smelter exceeded government fluoride emission standards. ⁵¹

Rural residents in China's Inner Mongolia also felt the impacts of the Chinese aluminum industry. A few years after one smelter started operating, herders in the surrounding area said their sheep began falling sick, with jaws so painful that they could not eat, and thousands soon died, according to the Washington Post. When the herders complained, the government arrested five of their leaders, forcing others to resettle in the nearby city of Holingol and demolishing their rural homes. The Huomei Hongjun plant in Holingol was one of the largest aluminum smelters in the world. Its coal-fired power plant was fined by the Chinese Environmental Protection Ministry in 2011 for excessive sulfur dioxide emissions and for falsifying emissions data. A government document obtained by Ceng Jing Cao Yuan, a local advocacy group, showed that more than 12,000 sheep had died and 23,000 fell sick in seven villages and two farms near the factory between 2008 and 2009. The government document said the probable cause was high levels of fluoride in their bones and in the grasslands, but tests on herders had showed no ill effects in humans. The authenticity of the document, which carried no official stamp, could not be independently verified, but the symptoms described by the herders were consistent with fluoride poisoning, the Washington Post reported. ⁵²

Ends of the Earth

Fluoride emission problems from aluminum smelters can now be found in Third World nations around the globe. Bauxite deposits are commonly found in tropical areas, and to

improve financial logistics, alumina refineries and aluminum smelters are sometimes built near the bauxite mines. But with more knowledge publicly available about the hazards of fluoride emissions, proposals to build such large projects are sometimes met with opposition by local or international environmental interests. In September 2002, the Gulf International Investment Group, including Dubai Aluminum, announced plans to spend \$2 billion building a 500,000 ton-per-year aluminum smelter in Sarawak, a Malaysian state on the island of Borneo. The new smelter would use power from the proposed 2,400-megawatt Bakun hydroelectric dam. Dubai Aluminum was one of the top 10 aluminum producers in the world. ⁵³ In August 2002, Friends of the Earth-Malaysia issued a press release protesting construction of the Sarawak aluminum smelter and associated hydroelectric dam. The organization claimed major environmental and health problems could result from fluoride emissions by the plant, including skeletal fluorosis and other long-term health effects to both wildlife and humans. ⁵⁴

In October 2002, salmon farmers in southern Chile threatened to shut down operations and move away if the Canadian mining company Noranda built its \$2.75 billion, 440,000 ton-per-year Alumysa aluminum smelter there. The salmon farmers claimed the plant would emit 300,000 tons of fluoride per year and damage their fishing industry. Noranda responded by noting that aluminum smelters in Norway and Iceland did not harm nearby salmon farms there. The new Chile facility would include 758 megawatts of hydroelectric capacity. ⁵⁵ The Aysén region of Chile was promoted by opponents to the smelter as one of the three least contaminated areas on the planet, and residents of the region declared Aysén a "Life Reserve." The proposal encountered significant opposition by a large number of Chilean and international organizations concerned about the project's impacts on the fragile ecosystem and the people of Chacabuco Bay and Patagonia. ⁵⁶

At the opposite end of the Earth from Patagonia, an aluminum industry has developed on Iceland to take advantage of hydroelectric and geothermal energy. Iceland's first aluminum smelter was built in the 1960s. Two smelters were operating near Reykjavik by 2007, and a new smelter proposed by Alcoa would utilize power from the \$3 billion Karahnjukar Hydropower Project. Plans called for eight more hydropower and geothermal plants, two more aluminum smelters and expansion of an existing smelter, bringing Iceland's aluminum smelting capacity to 1.6 million tons per year. "If all of these projects get through, then it's a total environmental apocalypse for the Icelandic highlands – they'll have developed every single major glacial river and geothermal field for heavy industry," Saving Iceland organizer Olafur Pall Sigurdsson told the New York Times in 2007. "It is a very rare nature that we are the guardians of, and we are squandering it." The new smelters would require about eight times the amount of electricity used by all of Iceland's domestic consumers. Hjorleifur Guttormsson, Iceland's energy and industry minister from 1980 to 1985, said he was concerned about sulfur dioxide, hydrogen fluoride and other chemical emissions. ⁵⁷

In August 2006, the pollution control scrubbers at Century Aluminum's 280,000 ton-peryear Nordural aluminum smelter at Hvalfjordur in West Iceland stopped working for about 20 hours. The plant was expanding capacity at the time of the accident, and large amounts of fluoride were emitted in the fjord area where the Norwegian metallurgical company Elkem also operated a steel alloy factory. After farmers complained about impacts to their livestock, tests found that the heads of affected sheep contained fluoride levels up to 1,300 ppm and 1,400 ppm when the normal level was 300 ppm to 400 ppm. Across the fjord at another farm, tests of horses showed fluoride levels about four times above expected baseline levels. The Icelandic government tended to take the side of the aluminum producers, according to the Saving Iceland website - in spring 2011, the government's food and veterinary authority said the affected horses were overfed. That same year, plans were announced to expand industry at Hvalfjordur. In October 2012, high levels of fluoride were reported in hay grown on three farms around Alcoa's Fjardaal aluminum smelter in East Iceland. In response, Alcoa announced that their pollution control technology had failed sometime during the summer. Tests on the hay in January 2013 showed fluoride levels below the maximum limit for cows, goats, sheep and dairy cattle. Two of the 17 samples were above the acceptable limit for milking cows for human consumption, but the farm only had horses. ⁵⁸

Rationale for regulation

E.R. DeOng first described plant injury near a U.S. aluminum smelter in 1946. From that time until Clinton Carlson's 1978 Ph.D. dissertation on the impacts of fluoride from the Anaconda Aluminum Co. smelter in Columbia Falls on nearby forests, nearly all studies of the effects of fluoride on vegetation focused on describing visible injuries, the geographical extent of the injury, and the morphological or physiological and biochemical effects. "Little work has been published in which fluoride-caused losses have been quantified," Carlson said. In 1974, T.D. Waddel, at the U.S. Environmental Protection Agency, stated that "given the paucity of knowledge in the literature on pollutant-yield relationships, many of the damage factors were probably guesses." Carlson wanted those estimates, or "best guesses," improved. "Information of this type is politically important and is relevant to land managers and others responsible for administration of pollution-damaged public lands," Carlson said. "Also, the data may be relevant to standards set to protect human health and welfare." ⁵⁹

In 1969, the British chemist R.Y. Eagers published a book on the toxic properties of inorganic fluorine compounds. Since the beginning of the 20th century, the use of

fluorine compounds had increased significantly, particularly in industry. Fluorine was estimated to be the 13th most abundant element in the Earth's crust, but it rarely existed in a pure gaseous form in nature. In the mid-1930s, a study of injuries to animals and vegetation by dusts or gases emitted by industry identified the sources as six phosphate plants, six chemical plants, five aluminum smelters and one brickworks plant. Another study found 71 cases of injury to man and livestock between 1935 and 1957, of which 22 were related to aluminum smelters and 18 to chemical plants. One of the early studies showed that the atmosphere 200 yards from an aluminum plant contained 0.22 milligrams of fluorine per cubic meter, while at a distance of one mile away the atmosphere contained 0.042 milligrams per cubic meter. The soil near this aluminum plant contained 66 ppm of fluorine. Furthermore, the mandibles of sheep near this aluminum plant contained fluorine at levels of 17,000 ppm, while the radii and ulnae of the cattle contained fluorine at levels of 9,600 ppm.⁶⁰

Eagers used the term chronic fluorosis for injuries to animals or plants caused by exposure to fluoride through inhalation or surface exposure over a long period of time. Chronic fluorosis developed slowly as the fluorine was retained in the skeletal system – it was possible for animals to tolerate for short periods of time levels of fluorine that would be hazardous over a longer period of time, Eagers found. Typically, animals exposed to fluorides over a long period of time developed mottled and stained teeth with excessive wearing, and eventually they showed signs of lameness and loss of appetite. In 1955, a National Research Council committee proposed safe ranges of fluoride for various farm animals. By 1969, most investigators concluded that animals were not injured directly by fluoride in the atmosphere but by fluoride that was deposited on herbage eaten by animals. This fluoride dusting could be removed by washing the leaves. Plants, on the other hand, were less affected by fluoride particulates landing on the surface of the plant as by gaseous fluorine compounds that entered the stomata openings in the leaves. Over time, the leaves accumulated gaseous atmospheric fluoride. Older needles of ponderosa pines were less susceptible than younger needles. Plants could also absorb fluoride through the soil, with identical injuries. Eagers reported. ⁶¹

According to a U.S. Organization for Economic Cooperation and Development report in 1977 on air pollution in the primary aluminum industry, fluoride emissions did not pose a significant human health hazard. Severe effects had only been observed on humans after long-term occupational exposure to high levels, such as in the mining of cryolite or phosphate. According to the OECD report, the National Academy of Sciences found that community airborne fluoride had caused an adverse effect on humans in the very few instances where humans were in the immediate vicinity of fluoride-emitting industries.

The threshold limits for adverse human effects by hydrogen fluoride was 2.45 milligrams per cubic meter and by particulate fluoride was 2.5 milligrams per cubic meter. Among animals, dairy cattle were found to be the most sensitive. Evidence suggested that no adverse effects were found in dairy cattle if the concentration of fluoride in feed or forage was less than 40 ppm over a full year, 60 ppm for two consecutive months or 80 ppm for more than one month. Airborne fluoride was found to adversely affect the growth, development and yield of vegetation depending on a variety of factors, including the species of plant, the variety within a species, the stage of a plant's life cycle, temporal patterns of exposure and the presence of other pollutants.⁶²

According to the 1977 OECD report, results from laboratory experiments on plants and from data collected on vegetation near aluminum smelters were believed to be too complicated to fully understand. Generally it was felt that the lack of precision created difficulties in setting emission standards, so the report suggested guidelines be set more stringently to be on the safe side. The types of fluoride emission standards adopted worldwide varied according to several factors: 1) national versus local determination and enforcement; 2) standards that were meant to be objectives rather than minimum or maximum limits; 3) legislative wording intended to protect human health, to conserve natural resources or to protect the living environment; 4) variance in how scientific or medical information was interpreted; and 5) description of pollution control technology as best available or best practicable. In the U.S., national standards for fluoride emissions were based on the "best control technology" that was "taking into account cost" what had been "adequately demonstrated." State governments were expected to develop and set fluoride emission standards that could in some cases be stricter than federal standards. ⁶³

Regulating agencies were well aware of the harmful impacts of aluminum smelter emissions, but establishing rules and regulations came only after lengthy studies and discussion. In 1971, the Singmaster & Breyer firm of New York completed a report for the Environmental Protection Agency on air pollution control in the primary aluminum industry. "Solids and gaseous fluoride effluents from an aluminum reduction cell constitute the most serious aspects of the air pollution abatement problem," the report said. All three types of aluminum reduction cells – prebake, vertical-stud Soderberg and horizontal-stud Soderberg – emitted about the same amount of fluoride as effluents, the report stated – about 23 pounds of fluoride emitted per 1,000 pounds of aluminum metal produced. Fluoride losses in reduction pots resulted from cryolite absorption into cell linings and dusting on the outside of the cell. About three-quarters of all fluoride losses occurred because of dusting, which occurred when the cell crust was "worked," or broken, to add alumina or cryolite to the molten bath. Fluoride emissions from a typical reduction cell in the U.S. could increase from 15 pounds per 1,000 pounds of aluminum produced to 53 pounds when the crust was worked. The crust on Soderberg cells was routinely broken by a pneumatic crustbreaker, which could typically last about five minutes. ⁶⁴

About one-third of the fluorine content of cell emissions was in the form of particulate fluorides, some of which were large enough to be recovered by pollution control devices and returned to the cell, Singmaster & Breyer reported. Finer sizes were more difficult to remove from the emissions. Gaseous fluorides evolved directly from the cell or were formed by hydrolysis from other fluorine compounds. Gaseous fluorides could be completely removed by chemisorptions, such as adsorption in dry scrubbers, or by aqueous media. Other pollutants in air emissions from aluminum smelters included carbon or alumina dust from materials handling and preparation, and particulates and gases produced in anode bake furnaces or cast houses. Those emissions could be addressed by "housekeeping" and industrial hygiene procedures, the report said, but potline emissions "are the greatest quantity and potentially the most damaging." Most of a reduction cell's pot gas was carbon dioxide or carbon monoxide, with carbon dioxide making up 60% to 85%. Other compounds found in pot gases included sulfur dioxide, hydrogen sulfide, carbonyl sulfide, carbon disulfide, silicon tetrafluoride, hydrogen fluoride and water. Water vapor in the air that entered the cell provided the hydrogen needed to create hydrogen fluoride. Some of that hydrogen fluoride combined with particulates before leaving the cell. 65

Potline emissions for prebake cells typically included 30 pounds sulfur dioxide, 14 pounds gaseous fluorides and nine pounds solid fluorides per 1,000 pounds of aluminum metal produced, Singmaster & Breyer reported. Potline emissions from Soderberg cells, like the cells used at the AAC smelter in Columbia Falls, also contained volatilized hydrocarbons. Data from U.S. smelters using air pollution control systems in 1970 indicated that overall emission control efficiency was 73% for total solids, 66% for solid fluorides, 84% for hydrogen fluoride gas and 77% for total fluoride. That added up across the U.S. to 23,200 tons of fluoride emissions from potlines in 1970, including 10,200 tons of gaseous fluorides and 13,000 tons of particulate fluorides for 4 million tons of metal produced. Aluminum producers reported spending \$1.5 billion (in 2016 dollars) altogether on emission control equipment, of which \$1.4 billion went to potline emissions control. The industry as a whole spent about \$407 million per year on operating costs, including interest payments, taxes, insurance and depreciation. ⁶⁶

Singmaster & Breyer followed up in July 1973 with another report on air pollution control in the primary aluminum industry for the EPA. Principal authors John C. Russell and Dumont Rush described the importance of the "burner" placed in the pot gas flue pipe for a vertical-stud Soderberg type of reduction cell, like the ones used at the AAC

smelter in Columbia Falls. Unlike prebake cells, where carbon blocks to be used as anodes were baked ahead of time in a special oven, the baking of the carbon paste in the anode of a Soderberg reduction cell took place in the potroom while the cell operated, meaning hydrocarbon fumes and volatiles derived from the pitch binder in the carbon paste were constantly being released. Pot gas from vertical-stud Soderberg cells contained 3% hydrocarbons, primarily methane, ethane, propane, butane and related isomers. ⁶⁷ In the case of the reduction cells at the AAC plant in Columbia Falls, the burner was located at the base of the heavy gauge steel flue pipe mounted at each end of the anode. Once ignited, air entering the burner kept the flame going so long as combustible fumes, primarily carbon monoxide, were emitted from the reduction pot. Winds common to the enormous potrooms also could blow out the flame.

The "tarry" nature of the hydrocarbon effluents in pot gas could foul most air pollution control equipment, including wet scrubbers, because the "tarry" effluents did not dissolve in water. But a burner in the pot gas flue pipe could convert the tars to gaseous fractions that did not interfere with the operation of the plant's air pollution control equipment. "This system has the advantage of very low air dilution and sufficiently high hydrocarbon concentration that the effluent gases may be burned," the Singmaster & Breyer report stated. "Most of the hydrocarbons, both gaseous and particulate, and much of the carbon monoxide, is oxidized into carbon dioxide and water vapor. The effluent gas leaving the burner is low in hydrocarbons and may then be treated in a manner similar to prebake cell effluents." After combustion, pot gases from verticalstud Soderberg cells contained less than 0.1% hydrocarbons, indicating a 96.7% efficiency. On the other hand, the absence of a hood on vertical-stud Soderberg cells meant hydrocarbons were emitted to the potroom, creating secondary emissions. When a reduction cell was operating normally, the burners maintained a continuous flame, but irregularities in operation of the cell could cause a flame-out. Without automatic igniters or a satisfactory program for manual ignition, it was possible 5% to 10% of the burners in a potline could be out at any given time. Pot gas flue pipe burners could not be used with horizontal-stud Soderberg cells, the report noted. ⁶⁸

The Soderberg problem

By 1972, twenty of the 30 aluminum smelters in the U.S. used prebake reduction cells, four used vertical-stud Soderberg cells and six used horizontal-stud Soderberg cells. One of the conclusions the Singmaster & Breyer report in 1973 reached was that the addition of secondary treatment to primary treatment systems amounted to a small yet significant increase in overall control efficiency. This conclusion was even stronger for vertical-stud and horizontal-stud Soderberg cells, which could not be easily hooded. The efficiency for air pollutant control in primary emissions for vertical-stud Soderberg

ranged from 75% to 99% for particulate fluorides depending on the type of equipment, and from 93% to 99% for gaseous fluoride. But the overall control efficiency ranged from 78.8% to 78.7% because there was no secondary emission control. ⁶⁹ Primary emissions included pot gases and were directed by flue pipes and ducting to air pollution control equipment. Secondary emission control typically included rooftop control equipment that captured gases and particulates that left the reduction pots and entered the potrooms. The AAC plant in Columbia Falls installed dry scrubbers in the late 1970s to control fluoride in pot gas, but the plant never installed permanent secondary emissions control. A pilot program was unsuccessfully tried out in Potroom 8.

The Norwegian inventor Carl Wilhelm Soderberg patented the Soderberg anode for aluminum reduction cells in 1918. The invention was used in the aluminum industry since 1923 and was considered the first important breakthrough invention for the industry since the Hall-Heroult discovery in 1886. By 1923, prebaked anodes had been in use for about 40 years, but the problem of carbon consumption in the reduction process was more economically addressed by the Soderberg design, which created a continuous self-baking and monolithic anode. The heat from the electrolyte in the pot baked the anode to just the right consistency to conduct electricity for aluminum reduction. The main advantages of the vertical-stud Soderberg anode was the savings in capital, labor and energy required to manufacture prebaked anodes. The disadvantages of the vertical-stud Soderberg reduction cell included higher pot voltage and energy consumption, lower current efficiency, lower quality anodes, smaller pot size, and higher emissions of fluoride and polycyclic aromatic hydrocarbons.⁷⁰

An example of an aluminum smelter using Soderberg technology was the smelter plant in Chalmette, La., that Kaiser began operating in February 1951. This was Kaiser's first greensite project. For some time, the 270,000 ton-per-year plant was the largest in the world. Kaiser chose to install Soderberg cells, believing the technology was the most advanced at the time for lower operating costs and higher metal purity as compared to prebake cells. Alcoa and Reynolds were installing Soderberg cells at the time, and "notorious atmospheric emissions were not a concern at that time," George Binczewski reported in the Journal of Metals in 2002. To deal with the fumes, Kaiser initially built a 550-foot high smokestack in hopes of distributing the emissions in the upper atmosphere. In 1976, the company installed a \$32 million dry scrubber system to deal with the pot fumes.⁷¹

According to the EPA, 17 out of 23 primary aluminum smelting plants in the U.S. in 1997 used prebake technology rather than Soderberg. ⁷² Modifying Soderberg pots rather than replacing them with prebake pots was a long-held goal among global aluminum producers, especially as environmental regulations were further developed and

implemented. In September 2003, ENAL Newsletter published an editorial by Helge Forberg describing the economics of revamping Soderberg aluminum smelters. Despite low metal prices worldwide, the high cost of building new smelters made upgrading existing Soderberg pots economical. The debt service for a new smelter with the latest equipment ran to about 36% of total operating costs, a record high. Soderberg pots had claimed a good operating performance 50 years earlier, when vertical-stud Soderberg pots were introduced. That was a time of rising aluminum demand, and smelters with Soderberg pots had lower operating costs than the typical aluminum smelter with prebake pots, according to Forberg. Operation of the early Soderberg pot depended on the skill of the smelter's foremen and operators because the pots were generally operated manually. But because many plants were not run effectively, Soderberg pots acquired a bad image over the next 25 to 30 years, Forberg said.⁷³

During the 1970s and 1980s, many Soderberg pots were upgraded with Sumitomo technology, which improved performance, increased production and reduced emissions, according to Forberg. This upgrade helped to extend the economic life of older smelters and showed that revamping older Soderberg plants could make them competitive, assuming the aluminum plants had acceptable power contracts. Properly operated Soderberg pots with good gas skirts and Sumitomo-type bar breakers could record 95% gas collection. A pot equipped with point feeders could do even better. A new Soderberg smelter with a capacity ranging from 50,000 to 100,000 tons per year could be built without the severe cost penalty associated with a new prebake plant because the Soderberg plant would not need a baking furnace and rodding room to manufacture prebaked anodes, which could account for 20% of the cost of building a new prebake plant. "Additional cost savings could possibly be found by selecting a simplified plant layout similar to the Harvey Aluminum smelters built many years ago and by basing the operation on the purchase of anode paste," Forberg said. "However, it has to be recognized that the Soderberg pot has some inherent handicaps compared to a prebake pot." ⁷⁴ The AAC smelter in Columbia Falls was converted to Sumitomo technology in the 1970s.

Polycyclic aromatic hydrocarbons emitted by Soderberg anodes generally were considered markers for incomplete combustion and included a wide variety of chemicals, including acenaphthylene, pyrene and benzo(a)pyrene. Some polycyclic aromatic hydrocarbons were considered persistent, bioaccumulative and toxic and were suspected to cause cancer and developmental, reproductive and neurological impairment. "Once deposited to water and land, these compounds build up in organisms and magnify in concentration with each level of the food chain," the Western Airborne Contaminants Assessment Project reported in 2015. ⁷⁵ According to an article about innovative aluminum smelting technologies in the 2014 Journal of Occupational & Environmental Medicine, carbon paste briquettes for Soderberg anodes were made with twice the amount of coal tar pitch as used to manufacture prebake anodes. Soderberg anodes were difficult to enclose, and polycyclic aromatic hydrocarbon fumes from the self-baking anode escaped into the potroom, including benzo(a)pyrene, a suspected carcinogen. Soderberg anodes also had about 30% more electrical resistance than prebaked anodes, which reduced current efficiency. As a result, Soderberg technology was being replaced around the world.⁷⁶

Particulate pollution by aluminum plants that took place outside the potlines was typically caused by raw material unloading and handling. This included alumina, which arrived at aluminum smelters as a very fine white powder, and carbon materials destined for the smelter's carbon plant, typically coal or petroleum coke. Some plants that began as smelters ended up specializing in carbon products after the potlines shut down. By 2013, Alcoa's Lake Charles, La. plant was capable of producing up to 283,000 tons of calcined coke and 138,000 tons of carbon anodes per year. The plant, which Reynolds acquired from Conalco in the 1980s, employed 182 workers. Reynolds merged with Alcoa in 2000. ⁷⁷ On Jan. 13, 2015, it was reported that 27 Louisiana landowners had filed suit against Reynolds and Alcoa for coke dust emissions from the Lake Charles baked carbon anode plant. The plaintiffs alleged that nearby properties were damaged by "inordinate quantities" of coke dust landing on their homes and vehicles. The plaintiffs sought money to pay for a scientific investigation of the contamination, to clean the affected properties and to compensate the landowners for the "annoyance, discomfort and inconvenience" resulting from the emissions, as well damages up to \$74,500 apiece. Alcoa asked that the case be transferred to U.S. District Court. 78

Pollution control technologies

The 1973 the Singmaster & Breyer report for the EPA on air pollution control in the primary aluminum industry described the complex chemical process that takes place in a Hall-Heroult reduction cell. "Several theories have been proposed to account for the physical changes which occur during the electrolysis of aluminum oxide," the report said. "However, the high reactivity of the complex electrolyte combined with the high operating temperature make it difficult to determine experimentally which ions are present. Little is known about the exact reaction mechanism beyond that it is complex and is variable with both temperature and with the concentrations of the several bath constituents." The Singmaster & Breyer report focused on air pollution by fluoride emissions. "Cryolite bath is gradually lost from the reduction cell through absorption in lining materials, electrolysis and vaporization," the report said. "Although the quantity varies among aluminum reduction plants, about 20 to 50 pounds of cryolite must be added to the bath per 1,000 pounds of aluminum produced in order to make up for

these losses." The optimum ratio of sodium fluoride to aluminum fluoride in the bath for aluminum production was adjusted by plant workers to fall between 1.30 and 1.45, but sodium impurities in the alumina which was added to the cells could accumulate in the cell and react with the molten cryolite to form additional sodium fluoride and increase the bath ratio. Bath losses also occurred by vaporization in the cell and the reaction of cryolite with water that entered the cell. To maintain optimum bath ratio, plant workers periodically added aluminum fluoride. Approximate quantities of feed materials to Hall-Heroult cells to produce 1,000 pounds of aluminum in 1971 included 1,950 pounds of alumina, 44 pounds of bath, a mixture of cryolite, aluminum fluoride and fluorspar, 457 pounds of calcined coke, 138 pounds of pitch and 7 to 8 megawatthours of electrical power.⁷⁹

By 1981, pollution-control costs for the U.S. aluminum industry ran to about 3 to 4 cents per pound of aluminum produced, or about 5% of the list price. By comparison, pollution-control in the copper industry ran to about 20 cents per pound. All mining and mineral processing industries faced the same task of extracting small amounts of metal from large amounts of typically low-grade ore. Each stage of the process, from mining and ore processing to refining and smelting, affected the surrounding environment. Environmental degradation resulted from vast amounts of waste rock, effluent streams, gases and particulates. The most well-known pollution problem in the aluminum industry was atmospheric emissions from smelters. With no pollution control equipment in place, a theoretical 220,000 ton-per-year aluminum smelter could emit 40 to 60 pounds of fluorides and 112 pounds of complex fluoride compound particulates, such as sodium aluminum fluoride or calcium fluoride, per ton of aluminum produced. The same theoretical plant could emit 1% by volume carbon monoxide and carbon dioxide, from 5 to 47 ppm sulfur dioxide or sulfur trioxide and 5 ppm nitrous oxides. In general, prebake plants had less difficulty in controlling fluoride emissions. It was estimated that pollution controls added from 5% to 15% to the cost of operating a U.S. smelter.⁸⁰

Singmaster & Breyer described 21 types of air pollution control equipment in use or being considered for use to reduce primary and secondary emissions at aluminum smelters around the world. They included burners on flue pipes, incinerators, multiple cyclones, baghouse filtering, fluid-bed dry scrubbers, coated-filter dry scrubbers, injected-alumina dry scrubbers, dry electrostatic precipitators, wet electrostatic precipitators, spray towers, spray screens, high-pressure spray screens, wet centrifugal scrubbers, venturi scrubbers, chamber scrubbers, wet impingement scrubbers, crossflow packed-bed scrubbers, floating-bed "bouncing ball" scrubbers, sieve plate towers, self-induced spray "bubblers," and vertical-flow packed-bed scrubbers. Some of the air pollution control systems cited in the list were used in foreign smelters and not in the U.S. For vertical-stud Soderberg cells, the fluid-bed dry scrubber reportedly provided 98% efficiency for removal of particulates and 99% for removal of gaseous hydrogen fluoride. The venturi scrubber reportedly provided 96% and 99% respectively, and spray towers provided 75% and 99% respectively. The most widely used dust collection equipment in all industry was the dry cyclone. For some aluminum smelters, using multiple tube cyclones as a preliminary particulate removal stage provided an economic incentive – the return of fluoride to the reduction cells. ⁸¹ The AAC plant in Columbia Falls started out by using spray towers to remove fluoride from pot gas, then experimented with venturi equipment before settling on the fluid-bed dry scrubber system.

One of the most effective methods for removing dry particulates according to Singmaster & Breyer was baghouse filters, where fabric filters could collect 100% of particles with a median size of 0.5 microns. The caked-on particulates could be removed from the fabric by intermittent reversal of gas flow or by physical shaking. The only time baghouse filters were used for pot gas was after they passed through a dry scrubber. Dry scrubbers utilized the adsorption of hydrogen fluoride gas to alumina particles, with the reacted alumina being used in the reduction cells, thereby returning fluoride back to the reduction pots. The "chemisorption" process created aluminum fluoride and water. Three types of dry scrubbers were in use or being considered in 1973 – coated filter, injected alumina and fluid bed. In the Alcoa Method 398 fluid-bed dry scrubber, which was installed at the AAC smelter in Columbia Falls during the Sumitomo conversion in the 1970s, alumina was continuously fed into a reactor in amounts up to 100% of the entire needs of the smelter's potlines, and pot gas was run through the floating alumina. Virtually all of the pot gas particulates were trapped in the reactor, with fugitive particulate (primarily alumina) stopped by baghouse filters mounted above the reactor. The baghouse filters were cleaned intermittently by pulsed pneumatic shaking, with the caked-on material dropping back into the reactor.⁸²

In an electrostatic precipitator, pot gas entered a relatively large chamber where particulates collected on negatively-charged wires suspended in the hot gas stream and on grounded collector plates. The removal efficiency of electrostatic precipitators could be improved by raising the moisture content of the pot gas. The spray tower was one of the most common type of pollution control equipment used for pot gases and was initially used by the AAC plant in Columbia Falls when it started operating in 1955. Particulate removal efficiency was low, so spray towers typically were used with multiple cyclones. Water with lime was used to react with the acidic fluoride gas and particulates. Typically the "scrubber liquor" was then run through a treatment facility which precipitated the fluoride out as calcium fluoride and returned the liquor back to the spray tower. It was conceivable that a smelter located near an alumina refinery

could send specially treated scrubber liquor to the refinery. At the other extreme, some plants dumped their scrubber water into rivers and streams without treatment. The venturi scrubber, which was tried unsuccessfully at the AAC plant in Columbia Falls in the late 1960s and early 1970s, contained a reduced flow area (throat) in the main pot gas duct where the velocity of the pot gas was increased many times, followed by a diffuser section. Scrubbing "liquor" injected into the throat would mix with the pot gas as an extremely fine spray. Venturi scrubbers worked best if the entering pot gas was saturated with water vapor. ⁸³

Some air pollution problems at aluminum smelters were characterized as "housekeeping" matters, particularly ensuring that particulates weren't blown away by winds during raw material unloading and handling activities, or keeping the crusts on Soderberg pots "sealed" to prevent fugitive fluoride gas or particulate emissions. But for some industry leaders, government regulators and scientists, the air pollution problem was linked to design limitations for pots using the Hall-Heroult reduction process, and solutions typically involved tweaking the historic process, according to Singmaster & Breyer. In 1963, J.L. Henry used an experimental aluminum reduction cell to study the Hall-Heroult process. Henry reported that the fluoride content of reduction cell emissions could be reduced by increasing the ratio of sodium fluoride to aluminum fluoride in the bath and by reducing the operating temperature. But he noted that attaining this perfect condition was difficult to accomplish even in an experimental cell.

Singmaster & Breyer suggested cooler bath temperatures could reduce the amount of bath salts that would vaporize and be carried away in pot gases. The normal operating bath temperature was from 970 to 980 degrees Celsius, but this relatively low operating temperature was near the freezing point of the electrolyte. Therefore, cell operators needed to pay close attention to prevent the cell from becoming too cold and "mucking up." Some reduction cells were said to be "sick" and needed to operate at temperatures above 1,000 degrees Celsius, in which case they never crusted over. Computercontrolled reduction cells had a better chance of being operated at lower bath temperatures, Singmaster & Breyer suggested.⁸⁵ Better reduction pot management also could help reduce perfluorocarbon emissions – greenhouse gases with a high global warming potential. According to the 2003 World Resources Institute report, the EPA's recommendation for reducing perfluorocarbon emissions included 1) improving alumina feeding technology with computer-regulated point feeders; 2) training operators to minimize the frequency and duration of anode effects; 3) using computers to optimize reduction cell performance; and 4) measuring perfluorocarbon emissions and cell operating parameters to determine relationships between the two.⁸⁶

The primary goal of the 1973 Singmaster & Breyer report was to inform the EPA about pollution control alternatives, their effectiveness and costs. The firm used information that was supplied in confidence by aluminum producers, typical performance data from equipment manufacturers and published information from technical literature. Engineering analysis was applied to determine the current performance levels of air pollution controls. Systems analysis was applied to growth projections to determine future costs and potential levels of emissions. Based on a 74.3% overall emission control efficiency in 1971, Singmaster & Breyer analyzed the conditions necessary to achieve four levels of improvement: 1) raise overall efficiency to at least 80%; 2) apply best-demonstrated control technology on primary (pot gas) emissions only; 3) raise overall efficiency to at least 90%; and 4) apply best-demonstrated control technology to both primary and secondary (potroom) emissions. ⁸⁷

By 1971, U.S. aluminum producers had spent about \$1.4 billion (in 2016 dollars) on air pollution control equipment and were spending about \$379 million a year on operating costs, Singmaster & Breyer reported. To achieve 80% efficiency industry wide, they would need to spend \$2.9 billion on equipment and \$740 million per year on operating costs. To implement best-demonstrated control on primary emissions, they would need to spend \$3 billion on equipment and \$721 million on operating costs. To achieve 90% efficiency industry wide, they would need to spend \$3 billion on equipment and \$721 million on operating costs. To achieve 90% efficiency industry wide, they would need to spend \$4.4 billion on equipment and \$1.2 billion per year on operating costs. To implement best-demonstrated control on both primary and secondary emissions, they would need to spend \$5 billion on equipment and \$1.2 billion on operating costs. The costs were based on a smelter plant's total annual capacity – a plant that operated at reduced capacity could end up paying much more because the air pollution control equipment would be in place but not operating at its full capacity.⁸⁸

Singmaster & Breyer concluded that further research and development efforts could be most practically focused on 1) improving hooding and collection systems; 2) reducing the amount of emissions from the Hall-Heroult aluminum reduction process; 3) improving the performance of existing emissions control equipment; and 4) additional research in fundamental research on pollution abatement technology. "The effective management of air pollution abatement in connection with the production of aluminum requires reliable information on the effluents and emissions of the plant and the effects of the plant emissions at locations outside the plant property," the report said. That meant good source sampling in the plant and ambient air sampling outside the plant. ⁸⁹

Effective pollution control depended on monitoring emissions. One of the difficulties in sampling a smelter's emissions was the large-volume, low-velocity air flows in secondary emissions, which escaped through doorways and rooftops, Singmaster & Breyer noted.

Sampling also needed to distinguish between particulate and gaseous fluorides, a task made more complicated by the high reactivity of gaseous fluorine compounds. In aluminum smelters where pot gases were collected in one location for air pollution control, sampling was not as difficult. But sampling needed to be conducted over a long enough time period to ensure the collected sample was representative of long-time average conditions and not atypical because of disturbances – such as pot working or anode effects. Three hours to three days of continuous or frequent sampling in one location was reported to be the industry practice. For secondary emissions escaping from potrooms, the pollutant accounted for only about 5% of the amount found in primary pot gas emissions. A possible solution to the difficulty of sampling secondary emissions was to conduct long-term sampling at multiple locations along a 1,200 foot long potroom. In 2016 dollars, it could cost \$217,000 to provide sampling platforms along the full length of a single potroom rooftop, Singmaster & Breyer noted. ⁹⁰

A four-man team of technicians, analysts and a supervisory engineer could cost \$326,000 per year to monitor an entire plant. One smelter plant reported spending about \$5,400 per year to replace equipment and \$108,000 per year on research by university or industrial research organizations. Much larger corporations with several aluminum plants maintained large central research and development facilities. Singmaster & Breyer concluded that the addition of secondary treatment to primary treatment systems amounted to a small, yet significant, increase in overall control efficiency. This conclusion was even stronger for vertical-stud and horizontal-stud Soderberg cells, which could not be easily hooded. The efficiency for air pollutant control in primary emissions for vertical-stud Soderberg plants ranged from 75% to 99% for particulate fluorides depending on the type of equipment, and from 93% to 99% for gaseous fluoride. But the overall control efficiency ranged from 78.8% to 78.7% at Soderberg plants with no secondary emission control. ⁹¹

The ultimate goal in the aluminum industry has been to find a completely new way to reduce alumina to aluminum. In a 1999 journal article, Barry Welch, a professor at the University of Auckland, New Zealand, reviewed alternative processes for aluminum smelting. The driving factors included reducing costs for electrical power, the initial costs of building reduction cells and environmental considerations. The latter included compliance costs for fluoride emissions then and greenhouse gases in the future. Environmental compliance for fluoride emissions could amount to about 10% of the cost of metal production. Welch found that past analysis and discussion of smelting alternatives had demonstrated that fundamental energy requirements were the same for all options because they all started with the same basic ingredient, alumina, and finished with the same product, molten aluminum metal. The basic ingredients for all the alternative processes he looked at included alumina, electrical power, carbon and

some recyclable chemicals. The output was aluminum and carbon oxides. After reviewing several technologies of the future, Welch noted that the four key elements to a successful alternative process included a satisfactory chemistry path, a suitable and practical operable reactor system, a suitable material to construct reactors and electrodes, and the ability to meet stringent environmental standards without driving up costs. He concluded that the economic gains of these alternatives were not likely to be dramatic, and the development costs would be considerable – "so much so that it is impractical for any one company to do it alone." ⁹²

Emission problems became significant in the world as aluminum plants proliferated and grew in size. The U.S. aluminum industry started as a small plant on Smallman Street in Pittsburgh in 1888 and increased steadily under Alcoa until World War II, at which point the federal government got involved and built dozens of alumina refineries and aluminum smelters. Many of those war-time plants continued operating after the war, and more plants were built as new companies entered the industry. To industry officials and residents who lived near aluminum smelters, it was no secret that the plants emitted hazardous chemicals that affected the surrounding environment. But common law nuisance complaints were not effective in preventing wealthy and powerful companies from damaging neighboring properties or federal lands. State and federal regulations were needed, but establishing those regulations and enforcing them would first require abundant scientific proof. In some cases, the aluminum industry employed their own scientists to establish doubt in academic or government research, but oftentimes the evidence was too obvious, and companies were forced to pay damages and come up with better emission control measures.

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