

Review

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Review

Reconciling Environmental Safety and Economic Feasibility: A Review of Soil Fluorine Management Strategies in South Korea

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Abstract: Soil naturally contains fluorine, but concerns arise when its levels or bioavailability are increased by human activities or specific compounds. South Korea faces challenges in regulating soil fluorine pollution despite implementing stricter total fluorine standards than many developed countries. Moreover, these regulations can be economically unfeasible because of the naturally high background levels of fluorine in the soil in Korea. This review explores the global landscape of soil fluorine management strategies and examines potential solutions that reconcile environmental protection with economic concerns. We recommend a shift towards regulating specific and hazardous fluorine compounds rather than total content, and prioritizing remediation efforts based on assessments of bioavailable fluorine. This approach may help Korea establish a more effective and sustainable strategy for managing soil fluorine pollution.

Keywords: fluorine; pollution; bioavailability; soil management standards

1. Introduction

Fluorine is the 13th most abundant element in the Earth's crust and exhibits high reactivity and electronegativity [1,2]. Fluorine containing minerals include fluorite (CaF_2), cryolite (Na_3AlF_6), apatite ($\text{Ca}_5(\text{PO}_4)_3(\text{OH},\text{F},\text{Cl})$), topaz ($\text{Al}_2(\text{SO}_4)\text{F}_2$), amphibole species, and mica species. Griceite, an extremely rare mineral, contains 73% fluorine [3].

Natural fluorine in minerals is mostly insoluble in water, very stable with minimal chemical reactivity, and has virtually no effect on the human body or ecosystems. Fluorine in soil can enter the atmosphere or water bodies through natural pathways, such as weathering, volcanic activity, marine aerosol emissions, and anthropogenic pathways, including the production and application of phosphate fertilizers, brickmaking, coal combustion, and aluminum smelting [4].

In trace amounts, fluorine plays an important role in teeth and gum health. It prevents tooth dissolution and inhibits bacterial growth in the oral cavity, thereby preventing periodontal diseases or cavities. Fluoride also aids in mineral deposition in teeth, and promoting re-mineralization processes [5]. However, consuming fluorine-contaminated water continuously and repeatedly over an extended period, can cause white spots on the teeth or deform the skeleton. The maximum permissible limit for fluoride in drinking water for human consumption is 1.5 ppm [6]; therefore, fluorine levels must be checked before using groundwater for drinking purposes.

In September 2023, the Korean government proposed to the Ministry of Environment amending soil fluorine management standards. The goal was to align these standards in line with international practices while also protecting public health and ecosystems. This amendment could potentially lead to harsher regulations compared to some developed countries. However, South Korea faces unique challenges due to the **naturally high background levels of fluorine in its soil**. The Korean Ministry of Environment, in collaboration with the Korean National Institute of Environmental Research, is currently conducting research to identify the need to adjust soil fluorine remediation standards and address any potential inconsistencies. However, this proposal has sparked debate among academics

and soil remediation companies, raising concerns about relaxing regulations in the absence of a comprehensive scientific awareness of the specific hazards posed by naturally occurring, less bioavailable forms of fluoride in Korea. The basis of this study is their query – "Is easing soil fluorine contamination standards truly in the best interests of the Korean people?".

To address this question, we first reviewed the current literature to explore the sources and behavior of fluorine in the soil. Subsequently, we summarized the current challenges in managing soil fluorine in Korea. Next, we examined the soil fluorine content regulation strategies adopted by various countries. Based on this comprehensive study, we offer perspectives on soil fluorine contamination management in Korea.

2. Origin and Sources of Fluorine

2.1. Natural Sources

Although fluorine is present in some rock-forming minerals, such as fluorite and apatite, it is more abundant within the Earth's lithosphere in hydroxysilicate minerals. In these minerals, fluorine substitutes for hydroxyl (OH) sites within the crystal lattice structure [4]. Fluorine is naturally derived from rock-forming minerals through weathering processes, wind-blown dust, marine-derived components, natural biomass burning, and volcanic activity.

2.1.1. Weathering

Weathering is a major natural pathway for fluorine in soil profiles [7,8]. Weathering is very complicated as it is governed by several intricate processes, such as adsorption-desorption and dissolution-precipitation. As granite massifs break down, fluorine is readily released at the onset of the weathering process [9]; however, fluorine content increases slightly in the uppermost layer of the weathered profile. While the fluorine in apatite remains stable, the fluorine from mica swiftly leaches out. Fluorite, if present, dissolves slowly due to the movement of water. Soil profiles typically exhibit a decrease in fluorine content as the distance from the source rock increases [10], with the majority of the fluorine in clastic sedimentary rocks found in their micaceous components [11]. Specifically, argillic limestone has an average fluorine content of 200 ppm, whereas its micaceous residue averages 800 ppm [11].

Fluorine-to-OH exchange in clay minerals is influenced by various factors, including fluorine concentration, circulating water pH, and specific clay minerals [12]. For example, dioctahedral and trioctahedral illite can adsorb fluorine by OH-to-F replacements. Fluorine is readily adsorbed onto clay structures in acidic environments, whereas it is readily desorbed in alkaline environments [12]. Rock weathering releases fluorine, some of which is adsorbed by clay in the surrounding water [13]. The overall soil fluorine content varies according to the composition and fluorine levels of the parent rocks, with an average fluorine content in the range of 90–980 mg/kg [14]. Recent studies have reported fluorine content of 321 mg/kg in soils worldwide and 557 mg/kg in the continental crust [15,16].

Organic matter decomposition is a key source of fluorine in warm and humid climate soils. Fluorine can easily leach away in acidic soils, whereas it accumulates in organic matter, which hinders leaching. Similarly, in permafrost regions, decomposing plants release fluorine into the water, exhibiting comparable behavior. Climate also influences fluorine mobility. The higher calcium content in arid regions restricts the movement of fluorine, while in semi-arid areas, such as India, surface and groundwater chemistry dictate fluorine migration. The dry season limits fluorine movement owing to high calcium and low dissolved solids. In contrast, the lower calcium and higher ionic strength in the water post-monsoon season enhance mobility [17,18].

2.1.2. Volcanic Activity

Volcanic activity emits considerable amounts of fluorine into the environment, primarily as hydrogen fluoride (HF) gas [15,19]. The majority of the fluorine (>90 %) is released during inactive periods (degassing) rather than during eruptions [20]. Volcanic ash traps and transports fluorine,

impacting areas beyond the immediate eruption zone. The effect of fluorine-rich volcanic emissions on drinking water and food sources is a major concern. Fluorine dissolves from ash in water and soil, potentially exceeding the permissible limits for human and animal consumption [21,22]. Animals grazing on fluorine-contaminated vegetation can develop chronic fluorosis, a debilitating condition affecting the bones and teeth [23,24].

Historical accounts also detail the devastating impact of volcanic fluorine on livestock populations, such as the mass die-off that occurred following the 1783–1784 Laki eruption in Iceland, which released an estimated 7 million tons of HF, resulting in widespread fluorosis and crop failure due to HF phytotoxicity [25]. Volcanic fluorine enrichment has also been documented in surface and groundwater sources [26] where high levels of soluble fluorine in volcanic ash leachate contaminate water supplies. Human populations living near volcanoes often depend on fluorine-enriched water sources, which leads to dental fluorosis [27].

Volcanoes have been suggested as the primary source of atmospheric fluorine, with estimates ranging from 0.06 to 8.6 million tons annually [2]. However, recent studies suggest that these estimates may have been exaggerated. Although volcanic fluorine emissions are considerable, they may not be as high as previously thought. In addition, although several researchers have suggested that we need to prepare for probable volcanic eruptions around the Mount Baekdu region in the near future [28], almost no marked volcanic activity has occurred on the Korean Peninsula in the last 1000 years.

2.1.3. Marine-Derived Components

Marine aerosol and spray contributions to atmospheric fluorine and its geochemical cycle have been debated. Friend [29] hypothesized a significant marine flow of fluorine to the atmosphere, estimated at 0.4 to 1 Mt annually and second only to volcanic emissions. Taverner and Clark [2] offered estimates between 1 and 2 Mt, while other authors predicted a smaller flux of 20,000 t. The average seawater fluorine level is 1.3 mg/L; thus, marine-derived fluorine is considered crucial in the hydrogeochemical fluorine cycle. Sugawara [30] and Carpenter [31] suggested that seawater is a major source of fluorine in global precipitation. Fluorine emissions from seawater include gaseous HF, with F/Cl ratios in precipitation being 10–1000 times higher than those in seawater [31,32]. However, Wilkniss and Bressan [33] discovered no preferential fluorine enrichment in marine aerosols and ascribed elevated fluorine levels to dust content. Barnard and Nordstrom [34] did not find a marked difference in fluorine content between coastal and inland rainfall, indicating anthropogenic fluorine sources in rainwater. De Angelis and Legrand [35] and Saether et al. [36] reported minimal marine-derived fluorine in ice and precipitation samples, respectively.

In contrast, Linder and Frysinger [37] and Lewandowska et al. [38] hypothesized that coastal rainfall and marine-derived aerosols are enriched in fluorine, which correlates with the Na content. Some areas—such as mid-Wales and coastal soils in Victoria, Australia—show fluorine enrichment, which might be attributed to marine sources [39]. Overall, despite contradictory evidence, marine-derived fluorine contributions to the atmosphere seem to be minimal when compared to volcanic and anthropogenic sources. Although seawater fluorine may influence coastal rainfall, runoff, and soil chemistry, its impact is likely limited to regions 10–20 km inland.

2.1.4. Other Minor Sources

Fluorine release into the atmosphere is not solely attributed to major natural sources, such as volcanic eruptions and marine-derived components; other sources also play a significant role. Weinstein [3] has suggested that wind-blown soil contributes substantially to the atmospheric fluorine content. Approximately 6,000 tons of fluorine are added to the atmosphere annually in the USA owing to the wind-induced removal of ~30 million tons of soil. Analytical data from ice cores in Greenland [35] and the Alps [40] indicate that wind-blown dust originating from soil was the primary source of atmospherically deposited fluorine before 1930. Despite anthropogenic activities dominating fluorine sources from the 1930s to 1980, wind-blown dust still accounted for $18 \pm 2\%$ of the fluorine in ice cores from the Alps between 1980 and 2000.

Biomass burning, both from natural processes or human activities such as agricultural practices and accidental fires, also contributes to atmospheric fluorine release. Although the majority of biomass fires are anthropogenic, natural fires—although less frequent—can destroy large areas of vegetation. De Angelis and Legrand [35] found elevated fluorine concentrations from biomass burning at high latitudes such as the Greenland ice caps, whereas Lewandowska et al. [38] identified biomass burning as a fluorine source in PM10 aerosols in the Baltic Sea area of Poland. Jayarathne et al. [41] investigated the fluorine emissions from biomass burning and found considerable amounts of fine particulates (PM2.5) containing fluorine; however, the degree of fluorine release varied with plant type and geographic distribution. Additionally, these authors estimated an annual release of 76,000 tons of fluorine from biomass burning, which is comparable to that from coal combustion. These minor sources, although individually smaller than major sources, collectively contribute substantially to atmospheric fluorine levels, highlighting the diverse nature of fluorine emission sources and their environmental impacts.

2.2. Anthropogenic Sources

Fluorine is also derived from anthropogenic sources. Emissions from industries—such as semiconductor manufacturing, steel production, aluminum smelting, and the production of glass, bricks, phosphate fertilizers, and items through electroplating processes—are important anthropogenic contributors to soil fluorine contamination [42].

2.2.1. Coal Combustion

Coal combustion has long been identified as a significant source of anthropogenic fluorine emissions [3]. Fluorine is considered one of the most hazardous substances released during coal combustion [43]. The average global fluorine content of coal is ~88 mg/kg [44], of which a substantial portion is released into the atmosphere during combustion. Chen et al. [45] suggest that nearly 80% of the fluorine present in coal is released in both gaseous and particulate forms at approximately 800 °C, with total release occurring at higher temperatures of 1100–1200 °C [46].

Several studies have highlighted the environmental consequences of fluorine emissions from coal combustion, including pollution, plant damage, and fluorosis in both animals and humans. For instance, emissions from coal-fired power stations have been identified as the single largest source of atmospheric fluorine in Australia, resulting in documented impacts on vegetation [47,48]. Similarly, fluorosis was observed in grazing animals near coal-fired power stations in the UK [48], and sheep near the Yatağan coal-fired power station in Turkey [49]. In Europe, studies have shown instances of fluorosis in wildlife, attributable to emissions from coal-fired power stations, particularly in regions such as the northwest Czech Republic [50,51].

As a major producer and consumer of coal, China faces acute challenges related to the fluorine emissions from coal combustion. Chinese coal contains an average fluorine content of 130 mg/kg, with much higher concentrations in certain regions, such as Guizhou Province [52,53]. The health effects of fluorine emissions are pronounced in China, where cases of fluorosis resulting from indoor coal combustion have been reported [52]. Dental and skeletal fluorosis have been documented in several Chinese provinces, affecting millions of people [54-56]. Exposure to fluorine is attributed to various sources, including burning coal in open stoves for food preservation and direct inhalation [52,54].

In addition to direct combustion emissions, coal waste is a considerable source of environmental fluorine contamination. Gao et al. [57] found high concentrations of fluorine in coal waste in China, with soils near waste sites having higher levels compared with background levels. The spontaneous combustion of coal spoil heaps adds to atmospheric fluorine pollution, potentially accounting for a substantial portion of fluorine emissions in China [57]. While most fluorine is released into the atmosphere during combustion, some is retained in the fly ash collected from power plant emissions. Although fly ash typically contains relatively low fluorine concentrations, its disposal in lagoons raises concerns regarding potential groundwater contamination [58].

Overall, coal combustion has emerged as a major anthropogenic source of atmospheric fluorine, with considerable environmental and health implications. Despite efforts to mitigate these emissions, the scale of fluorine release underscores the importance of continued research and regulatory measures to address this global challenge. South Korea currently operates 58 coal-fired power plants (36,868.7 MW) at 14 locations. As of 2022, coal-fired power generation will account for 39.7% of total power generation, making it the most significant source of electricity generation [59].

2.2.2. Brick and Ceramic Manufacturing

The production of construction materials, such as bricks and ceramics, has a hidden environmental cost associated with fluorine emissions [3,60]. Both brick and ceramic manufacturing involve the roasting of clays and clay-rich rocks at temperatures exceeding 900 °C [61]. These clays naturally contain fluorine originating from minerals such as micas [62]. During the roasting process, a considerable portion of fluorine is released into the atmosphere, primarily as HF and silicon tetrafluoride (SiF₄) [60]. Studies by Bonvicini et al. [62] and Xie et al. [63] found that > 80% of the fluorine present in the initial clay can be volatilized during firing.

Fluorine emissions have become a cause for concern in developing countries experiencing rapid urbanization. The increase in demand for bricks often leads to the expansion of inadequately regulated artisanal brickmaking practices [64,65]. These facilities, frequently located near urban areas [66], contribute notably to atmospheric fluorine pollution. Ahmed et al. [66] exemplified this problem in Peshawar, Pakistan, highlighting the presence of hundreds of brick kilns operating near the city. The resulting fluorine emissions were shown to be associated with damage observed in fruit trees and a substantial reduction in bean and maize crop yields [66,67].

The production volume and clay composition must be considered to accurately estimate the global impact of fluorine emissions from brickmaking. According to industry estimates [68], ~3 tons of clay are required to manufacture 1,000 bricks. Global brick production in 2010—concentrated in Asia with China, India, Pakistan, and Bangladesh leading the way—was estimated at 1,500 billion bricks, [69]. Assuming an average fluorine content of 500 mg/kg in clay (a range of 450–800 mg/kg; [70],[63]), and considering the 80% emission rate suggested by previous studies, we estimated an annual release of ~1.8 million tons of fluorine from brick production alone. This figure indicates the contribution of coal combustion, traditionally considered a major source of anthropogenic atmospheric fluorine [70].

The ceramic industry, which also relies on high-temperature clay firing to produce tiles and other products, faces similar challenges regarding fluorine emissions. Research by Bonvicini et al. [62] in Italy indicates significant fluorine emissions from ceramic tile production, although it likely represents only a portion of the impact of the global ceramic industry due to a lack of data on other ceramic products, such as sanitary ware. The data compiled from these references indicates that brick and ceramic manufacturing, particularly in developing countries, presents a significant risk of fluorine pollution in air which can have detrimental effects on vegetation and crop yields, as evidenced by a study in Pakistan [66]. The adoption of cleaner technologies in these industries is crucial for mitigating environmental impacts and protecting public health.

2.2.3. Fluorine Emissions from Aluminum Smelting

Aluminum production is based on the Hall–Héroult process, which releases fluorine into the atmosphere. Despite significant success in reducing fluorine emissions, ongoing monitoring and technological advancements are crucial for minimizing environmental and ecological impacts. These challenges and advancements in the management of fluorine emissions from the aluminum smelting industry have been extensively investigated [71-74].

During the electrolytic process that produces aluminum from aluminum oxide, fluorine is released in both gaseous (mainly HF) and particulate forms [74]. In the mid-20th century, the rapid growth in aluminum production led to severe fluorine pollution near smelters, resulting in ecological damage and health issues for livestock and wildlife [75-77]. The industry has responded by adopting cleaner technologies, particularly wet scrubbing systems, which have significantly reduced emissions

[78,79]. Estimates suggest that modern smelters emit as little as 15–30 kg of fluorine per ton of aluminum produced, with scrubbing further reducing this to <300 g per ton [71].

Despite these advances, fluorine emissions from aluminum smelters can still impact surrounding ecosystems. Rodriguez et al. [80,81] and Talovskaya et al. [81] documented elevated fluorine concentrations in vegetation and snowmelt water near smelters, highlighting their potential for long-distance transport. Hufschmidt et al. [82] and Kierdorf et al. [83] identified skeletal and dental fluorosis in kangaroos near an Australian smelter, indicating the potential health risks to wildlife. Global aluminum production is expected to reach 70.6 million tons by 2023 [74], and assuming most smelters limit fluorine emissions to 0.5–0.6 kg/ton of aluminum [71], an estimated annual release of 35,300–42,400 tons of fluorine is expected. However, data from World Aluminum [74] suggests slightly higher emission levels, potentially leading to an annual release of >45,000 tons [41].

2.2.4. Fluoride Release during Phosphate Fertilizer Production

The production of phosphoric acid—a key ingredient in fertilizers—poses a potential environmental threat due to fluoride emissions. Phosphoric acid is primarily produced from phosphate rock—a mineral rich in apatite—using wet and thermal processes [3,84]. The wet process accounts for 90% of global fertilizer production and utilizes sulfuric acid to extract phosphoric acid from rock [85]. Phosphate rocks naturally contain between 2–4% fluoride [86]. A large portion of this fluoride is converted to gaseous HF and SiF₄ during the wet process [3]. Although modern plants have implemented scrubbing systems to capture >99% of these fluoride-containing compounds [86], some emissions still occur. Studies on fertilizer plants in Brazil and Tunisia have emphasized this concern, with elevated fluoride concentrations reported in rainwater, groundwater, and nearby vegetation [87,88].

Another concern is the production of waste phosphogypsum, a byproduct of the wet process that contains calcium sulfate. Large quantities of phosphogypsum are stored in ponds, which can also contribute to fluoride contamination [84,86] as phosphogypsum contains fluoride, and the water in these ponds has high fluoride concentrations [3,85]. Leakage from ponds or windblown particles can further contribute to the dispersion of fluoride in the environment. Fluoride released from fertilizer production can travel through the air and contaminate the soil in surrounding areas. For example, studies in Turkey found elevated fluoride levels in soils near fertilizer production facilities, likely due to atmospheric transport from phosphogypsum storage [89].

Appropriate levels of fluoride are beneficial to dental health; however, excessive intake can lead to dental fluorosis, a condition that causes tooth discoloration and weakening. Tanouayi et al. [90] reported endemic dental fluorosis in a community near a phosphate mine in Togo, highlighting the potential health risks associated with fluoride contamination from mining activities. The global demand for phosphate rock for fertilizer production is considerable, with an estimated 263 million tons extracted in 2017 [91]. Assuming an average fluoride content of 3% in the rock and 10–15% volatilization during wet processing, a potential release of 0.694–1.04 million tons of fluoride could occur. However, these estimates only consider gaseous emissions from the acidification stage and do not account for particulates or releases from phosphogypsum storage. The existing literature suggests higher release quantities, with Jayarthne et al. [41] estimating combined gaseous and particulate fluoride emissions of 28,000 tons per year.

Although modern scrubbing systems have markedly reduced gaseous emissions, concerns remain regarding particulate matter, phosphogypsum storage, and the overall impact on local ecosystems and human health. Further research and stricter regulations are required to ensure sustainable practices in the fertilizer industry, minimize fluoride release, and safeguard the environment and public health.

2.2.5. Fluoride Contamination by Various Industrial Sources

The following industrial activities contribute to fluoride contamination of the environment.

Mining and Waste Management

Past and present fluorite mining activities pose a considerable threat in terms of fluoride contamination. In the United Kingdom, studies have documented extremely high soil and vegetation fluoride levels near abandoned fluorite mines and waste piles. Fuge and Andrews [92] reported soil fluoride concentrations >2% near mines in northern England and Wales. High fluoride accumulation is found in the vegetation at these sites, and concentrations up to 1% in plants from the Peak District of England were reported by Cooke et al. [93]. Researchers have linked elevated fluoride levels to dental fluorosis in grazing animals [94]. Similar environmental concerns exist around China-clay extraction sites in Cornwall, where fluoride-rich tailings contaminate nearby soils, plants, and waterways [92].

Fluoride in Steel Production

Fluorite is used as a flux in steelmaking, which releases fluoride as HF and SiF₄ gases [95]. Although a large portion of fluoride remains trapped in the slag by-product [96], historical accounts mention instances of fluoride-related environmental problems near steel plants, including animal fluorosis [97]. However, the implementation of pollution control technologies in modern steelmaking plants is likely to result in minimal atmospheric fluoride emissions [3].

Glass and Other Industries

While the glass and enamel industries have historically contributed to environmental fluoride contamination through HF emissions during production [95], the implementation of efficient scrubbing systems in developed countries has notably reduced this [3]. Additionally, the relatively small amount of fluorite used in these industries—compared to steelmaking—minimizes the overall impact [98].

Fluorocarbons and Emerging Sources

Hydrofluoric acid is used in various industries, including the production of fluorocarbons and semiconductors. Weinstein and Davidson [3] estimated that the semiconductor industry in the USA released ~32 tons of hydrofluoric acid annually during the 1990s. Furthermore, the petroleum industry uses it as a catalyst, and Lewandowska et al. [38] suggested that vehicle exhaust fumes may emit fluoride due to its presence in fuels.

2.2.6. Agricultural Sources

Agricultural practices—such as fertilizer application, sewage sludge disposal, and the use of certain agrochemicals—contribute to environmental fluoride contamination. Plants generally do not absorb fluoride from fertilizers, but it can potentially pose a threat to grazing animals and contaminate groundwater. Stricter regulations and monitoring of fluoride levels in agricultural inputs and practices are crucial for minimizing environmental fluoride contamination and safeguarding animal and human health.

Fluoride in Phosphate Fertilizers

Phosphate fertilizers—a major source of phosphorus for plants—are manufactured from phosphate rock that naturally contains fluoride. As discussed in the previous section, some fluoride is lost as gas during fertilizer production, however, some amount remains in the final product [86]. Studies in India revealed that between 3% and 75% of the original fluoride in phosphate rock remains in fertilizers [99]. Furthermore, practices such as recycling scrubbed HF gas back into the fertilizer during processing can further increase the fluoride content [42].

When applied to the soil, clay minerals and oxides strongly bind the fluoride from fertilizers and thus limit its uptake by plants. However, long-term application of phosphate fertilizers can result in fluoride accumulation in soil [100,101], raising concerns about the potential fluoride intake by grazing

animals through the ingestion of contaminated soil [42,101,102]. Considering the large amounts of phosphate rock used for fertilizer production [91] and the range of fluoride content in fertilizers (0.14–3.8 wt%), estimates suggest an annual addition of at least 2.3 million tons of fluoride to agricultural soils globally. Although fertilizer-derived fluoride is largely unavailable for plant uptake, soil ingestion poses a potential threat to grazing animals.

Soil Fluoride and Groundwater Contamination Risks

In some cases, fluoride from fertilizers can migrate from soil into groundwater, particularly under alkaline conditions [103]. Studies in India and Pakistan have reported elevated fluoride levels in groundwater from areas with extensive use of phosphate fertilizers [104,105].

Other Agricultural Fluoride Sources

Sewage sludge applied to agricultural land is another fluoride source. Sewage sludge can contain fluoride from various sources, including industrial wastewater, fluoridated drinking water, toothpaste, and medications [16]. Regulations typically limit the fluoride content of sludge applied to land (e.g., 200 mg/kg in the United Kingdom), but fluoride from sludge is likely to be more bioavailable than that from fertilizers [106].

Organofluorine-based agrochemicals, including insecticides, fungicides, and herbicides, are widely used in agriculture [107]. These compounds are persistent and resistant to degradation, raising concerns regarding their potential accumulation in soil and water resources [108]. Irrigation with fluorine-rich groundwater can also contribute to fluoride accumulation in plants, particularly via foliar uptake during overhead irrigation [109]. Botha et al. [110] suggested that fluoride-rich irrigation water may be partly responsible for livestock fluorosis in South Africa.

2.2.7. Urban Fluoride Emissions and Concerns

Many industrial activities that emit fluoride can considerably affect nearby urban areas. For example, artisanal brick production in Southeast Asia and aluminum smelting in Siberia are major sources of urban air pollution [65]. Coal and fuel combustion for domestic heating can also contribute to elevated fluoride levels in urban areas [38]. Vehicle emissions, possibly from fluoride-containing fuels and components, can also contribute [38]. The incineration of municipal solid waste releases considerable amounts of fluoride, primarily as HF. Although modern incinerators have emission control systems, the uncontrolled burning of waste can be a substantial source of HF and other fluoride-containing compounds in urban environments [3,111].

Airborne fluoride emissions from industry settle onto soil surfaces (dry deposition) or form acidic rain (wet deposition), and can impact the heath or nearby urban soils [112]. Plants play a further role in the transfer of fluoride to the soil by absorbing gaseous fluoride through their stomata (pores) or by taking it up from contaminated soil water [113]. While Perfluorinated compounds (PFCs) and chlorofluorocarbons (CFCs) are persistent environmental pollutants with a long history of industrial use, their potential contribution to soil fluoride levels in urban environments remains unclear and requires further investigation. [114].

2.2.8. Ubiquity and Persistence of Fluorinated Organic Compounds

The environment contains many fluorinated organic compounds, the majority of which are not naturally occurring [3]. These compounds are used in various applications, including agriculture, pharmaceuticals, and manufacturing. However, a major concern is the presence of perfluorinated compounds (PFCs), particularly those with long carbon chains. PFCs are resistant to degradation and can accumulate in the environment and biosphere [115]. Studies have shown their presence in humans and wildlife, with potential bioaccumulation and biomagnification effects [116]. PFC exposure has been related to a variety of health issues in humans, including cancer, thyroid disorders, and immune system dysfunction [117,118]. Some long-chain PFCs are classified as persistent organic pollutants due to their environmental persistence and adverse consequences.

Perfluoroalkyl and polyfluoroalkyl substances (PFAS) are a class of PFCs that are of major concern because of their potential toxicity [119,120]. While data are limited, estimates suggest that their environmental impact on overall fluoride levels may be negligible compared to that of natural sources [119,121]. This is likely because the strong carbon-fluorine bonds in PFAS make them resistant to degradation; however, their presence in the environment and potential health consequences warrant continued research and regulation.

Chlorofluorocarbons and Their Replacements

Once widely used in refrigeration, air conditioning, fire extinguishers, and aerosol propellants, chlorofluorocarbons (CFCs) have been discovered to be a major cause of ozone depletion [122,123]. Their production was phased out and replaced with hydrofluorocarbons (HFCs) and hydrochlorofluorocarbons (HCFCs), which do not harm the ozone layer [3]. While HFCs, HCFCs, and other fluorinated gases contribute significantly to greenhouse gas emissions [124,125], their potential breakdown products or atmospheric interactions might influence soil fluoride levels. Further research is needed to explore this connection.

Trifluoroacetic Acid: A Persistent Byproduct

The degradation of certain fluoride-containing compounds, including HFCs and HCFCs, generates trifluoroacetic acid (TFA). TFA is also released from industrial production processes, the burning of fluoropolymers, and household waste incineration [3,122,126]. TFA is highly stable in the environment and readily forms salts. Although low in remote areas, TFA concentrations are elevated in urban and industrial areas [3,126]. TFA is weakly bound to soil and has the potential to migrate into groundwater [127]. TFA accumulates in water bodies, such as salt lakes and oceans, due to evaporation; however, its presence in seawater at quantities greater than predicted from recent sources implies a natural source, possibly from hydrothermal vents [127].

2.2.9. Fluoride in Petroleum

Fluoride can occur in petroleum products because of its natural presence in crude oil or additives introduced during refining. This section explores the two main pathways for fluoride mobilization from petroleum sources—combustion and produced water [15]. Only the natural fluoride content of crude oil contributes to the additional anthropogenic mobilization of fluoride, as that added during refining originates from phosphate rock or mined fluoride and is already accounted for in environmental budgets. These estimates suggest that the average fluoride content of crude oil is very low (0.01 mg/kg) [128]. Considering global oil consumption data, the annual release of fluorine from petroleum combustion is estimated to be 0.000058 Tg [129]. Therefore, petroleum combustion appears to have minimal impact on the overall environmental fluoride budget.

Oil and gas production often involves the extraction of large volumes of water and hydrocarbons. These "produced waters" can contain large amounts of fluoride, with concentrations ranging from undetectable levels to over 190 mg/L [130,131]. Global oil production has increased steadily, leading to a corresponding increase in the amount of wastewater produced, with estimates suggesting that the water-to-oil ratio is ~3:1 globally [132]. Based on this ratio and the average fluoride concentration of 4 mg/L in produced water, these calculations indicate a small but increasing flux of fluoride associated with oil production. Estimates indicate that this flux increased from 0.04 Tg/yr in the early 1980s to 0.07 Tg/yr in 2018 [133]. While both petroleum combustion and produced water contribute to fluoride mobilization, their combined impact appears to be relatively less compared to that of natural sources. However, proper management and treatment of produced water is crucial to minimize potential environmental contamination from this source of fluoride.

3. Balancing Soil Fluorine Standards in Korea

The Soil Environment Conservation Act of 1995 serves as the cornerstone legislation governing the management of soil pollution in Korea [134]. Recognizing the importance of a healthy soil environment, the act identifies 24 soil pollutant types. Fluoride was included in 2002 alongside

organic pollutants like trichloroethylene, tetrachloroethylene, and inorganic nickel [135]. This inclusion reflected growing concerns about potential negative impacts of elevated soil fluorine levels. To establish these standards, the Korean Ministry of Environment developed a framework called Soil Worrysome Levels (SWL). This framework acknowledges that soil contamination often arises from secondary sources like air pollution, contaminated water, or various waste materials [135].

The current SWL for fluorine in South Korea is differentiated based on land use: 400 mg/kg for region I (croplands, rice paddies, residential areas, and schools) and region II (forests and playgrounds), and 800 mg/kg for region III (factories, gas stations, and roads) (Table 1). This tiered system reflects the potential health risks associated with varying exposure levels in different land use categories. However, implementing these standards presents significant challenges.

Table 1. Soil Worrysome Levels for fluorine in South Korea (unit: mg/kg).

Region	Fluorine content	Site description
I	400	Croplands, rice paddies, orchards, residences, and schools.
II	400	Forests, salt farms, playgrounds, and religious sites.
III	800	Factories, gas stations, roads, and military sites.

One major hurdle lies in the natural geological makeup of Korea. A large portion of the bedrock consists of granite [135], an igneous rock naturally rich in fluorine-containing minerals like feldspar and mica. This geology leads to widespread natural occurrence of fluoride in Korean soils. Studies by Lim et al. [136] supports this observation. Their evaluation of soil samples across 82 locations in Korea revealed an average fluoride concentration of 204.5 mg/kg (ranging from 15.3 to 504.8 mg/kg). This falls below the global average of 321 mg/kg. Notably, the average concentrations showed regional variations, with Region I, II, and III having averages of 229.6, 195.7, and 273.4 mg/kg, respectively. A nationwide soil survey conducted by the Korea Environment Corporation between 2012 and 2021 highlighted another challenge: widespread exceedance of SWL even in areas with naturally-occurring fluoride. The survey revealed that 1,337 (15.2%) of the 8,768 sampling points across Korea exceeded the SWL for Region I (400 mg/kg). This widespread exceedance, even in areas with natural sources, raises concerns about the practicality of the current regulations.

Furthermore, the high soil remediation costs associated with fluoride pose a major financial burden, particularly for housing development projects. The cost for fluoride-related soil purification from 2018 to 2022 amounted to 585.3 billion won (approximately 427 million U.S. dollars) in the metropolitan area of the capital Seoul alone [137]. Nationwide estimates are likely to be even higher. These substantial costs raise concerns about the economic feasibility of the current regulations.

While some experts claim that the current SWL (400 mg/kg) for sensitive areas such as croplands and residential areas is crucial for protecting public health and ecosystems by citing potential health risks associated with fluoride exposure, others advocate for a more nuanced approach. Proponents of a more nuanced approach emphasize that under natural conditions, the majority of fluorine in soil minerals is insoluble and very stable. Research directly linking health problems to stabilized soil-based fluorine compounds is limited. Although the presence of fluorine can help identify its source in the soil, the total amount of fluorine does not necessarily reflect its bioavailability [138]. However, research on the hazards of fluoride in air and water is well documented [92,139,140], raising the question of whether it is appropriate to relax SWL for fluorine. Examining other countries' strategies to manage soil fluorine contamination can provide valuable insights for establishing effective and practical regulations in Korea. This may entail amending current SML or implementing targeted remediation strategies based on factors such as bioavailability and land use.

4. Global Variations in Soil Fluorine Regulations

Table 2 summarizes soil quality guidelines for fluorine management from various sources. Before directly comparing Korean soil fluorine contamination standards with those of other countries, it is essential to recognize the significant variations in climatic conditions and geological characteristics across different regions. Our findings indicate that only a limited number of countries (at least 12) in North America, Europe, and Asia currently regulate soil fluorine as a potential contaminant (Table 2). These countries have established diverse soil fluorine management standards based on criteria such as climate, geology, and intended land use.

Soil quality guidelines for fluorine show wide variations, with values ranging from a minimum of 45 mg/kg for special areas with high biological value in Belgium to a maximum of 4,690 mg/kg for industrial areas in the same country (Table 2). This vast discrepancy highlights the significance of land use criteria when establishing soil fluorine management standards. Areas with potentially higher human exposure, such as residential or high-value ecological areas, often implement stricter limits.

Canada, which has a land area approximately 100 times that of South Korea, enforces a stricter limit (200 mg/kg) for agricultural land compared to Korea's current regulations [141]. However, Canada demonstrates flexibility by allowing a higher limit (2,000 mg/kg) for industrial and commercial lands. Similarly, Japan has a seemingly lenient standard of 4,000 mg/kg for soil fluorine [142]. However, it is important to consider that Japan enforces stricter regulation (0.8 mg/L) for fluoride in drinking water, than that recommended by the World Health Organization (1.5 mg/L) [143]. This example emphasizes how countries might prioritize regulations based on their specific environmental conditions and potential exposure pathways. Water-soluble fluoride content, in addition to the total amount of soil fluoride is the key factor influencing its mobility, plant uptake, potential toxicity, and risk of groundwater contamination [8]. The bioavailable fraction of soil fluorine is a more accurate predictor of potential environmental and human health risks.

The global trend in soil fluorine management indicates a move towards regulating specific fluoride compounds that pose clearly-defined risks. This approach prioritizes the management of synthetic or naturally occurring fluoride compounds that directly impact human health or ecosystems. The focus is on regulating these specific compounds, rather than the total soil fluorine content. The United States exemplifies this trend, where each of the climatically and geologically diverse 50 states has established independent soil pollution standards. However, a common thread across these standards is regulating the content of specific fluorine compounds, such as PFAS, which are detrimental to human health and ecosystems even at very low concentrations (section 2.2.8). Regulatory action is then taken through risk assessments when the concentration of these specific compounds exceeds established thresholds.

Table 2. Global soil quality guidelines.

No.	Country	Soil quality guidelines		References
		Land use	Concentration (mg/kg)	
1	Canada	Agriculture	200	CCME [141]
		Agriculture / Residential (Alberta)	200	
		Residential / Parkland	400	
		Commercial / Industrial	2000	
2	Australia	Industrial waste (Victoria)	450	EPA, Victoria [144]
3	Switzerland	All regions	400	Slooff et al. [145]
4		Regions with high clay content (>25%)	500	

	The Netherlands	Regions with very little or no clay content	175	
5	Austria	Agricultural / Residential (trigger value)	200	
		Agricultural / Residential (intervention value)	1000	
6	Belgium	Special areas with high biological value	45	
		Residential	3950	
		Industrial	4690	Carlton et al. [146]
7	The Czech Republic	Agricultural	500	
8	Italy	Residential / Public	100	
		Agricultural	2000	
9	Lithuania	Residential/ Recreational/ Agricultural	200	
10	Slovakia	Maximum allowable limits	1000	
		Value for decontamination measures	2000	
11	The United States (US)	Residential	469	USEPA [147]
12	Japan	All regions except agricultural	4000	Noh [142]

To summarize, soil fluorine management standards vary considerably across different countries due to diverse climatic conditions, geological makeup, and land-use considerations. While some countries have lenient total soil fluorine limits, they might compensate with stricter regulations for specific bioavailable fractions, water quality, or individual high-risk fluoride compounds. Understanding these nuances, as well as the global shift towards regulating specific high-risk fluoride compounds, is critical for establishing effective soil fluorine management strategies in South Korea.

5. Conclusions

In This study, we explored the sources, behavior, and management challenges associated with soil fluoride in Korea. We reviewed the current literature to understand the various sources of soil fluoride, including natural sources (weathering, volcanic activity) and anthropogenic sources (phosphate fertilizers, aluminum smelting). We also discussed the potential hazards of excessive fluoride intake, such as dental fluorosis and skeletal fluorosis.

An analysis of Korea's soil fluorine regulations highlights the complexities of balancing environmental protection with economic considerations. While some experts advocate for maintaining strict standards for total soil fluorine content, international practices and geological reality suggest the need for a more nuanced approach. First, in Korea, the naturally high background levels of fluorine in soil owing to the abundance of granite make adherence to stringent total content standards economically impractical. The considerable financial burden associated with remediating soil that exceeds these standards, even if naturally occurring, impedes development initiatives and raises concerns about the overall feasibility of these regulations. Second, global trends in soil fluorine management emphasize the regulation of specific fluorine compounds with proven risks. These typically include synthetic compounds or those exhibiting high bioavailability—such as PFAS—which have detrimental health and ecosystem effects even at low concentrations. Considering the above, South Korea may benefit from adopting a two-pronged strategy.

1. **Refine Standards:** Similar to the approach adopted in the United States for PFAS, soil quality standards should be revised for fluorine to focus on regulating specific, well-defined fluorine compounds with documented environmental and health risks. This shift would prioritize managing these harmful compounds.
2. **Bioavailability Assessments:** Implementing assessments to determine the bioavailability of soil-bound fluoride. This prioritizes remediation efforts in areas with high levels of bioavailable fluoride, which pose a greater risk of plant uptake, groundwater contamination, and potential health effects.

These approaches would enable South Korea to manage soil fluorine contamination more effectively, balancing the requirement to safeguard public health and the environment from an economic perspective. This would prioritize resources to address the most pressing concerns while minimizing excessive economic costs associated with naturally occurring, stable forms of fluorine in the soil.

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