EFFECTS OF FLUORIDE ION TOXICITY ON ANIMALS, PLANTS, AND SOIL HEALTH: A REVIEW

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ABSTRACT: Substantial multi-disciplinary efforts have been made to investigate the effects of environmental fluoride ion (F) pollution since the last century. The chronic ingestion of high doses of F may adversely affect human health by causing skeletal fluorosis, dental fluorosis, bone fractures, the formation of kidney stones, decreased birth rates, weakening of thyroid functionality, and impair intelligence, particularly in children. High concentrations of F in soil may seriously threaten the life of plants, devastate soil microbial activity, disrupt the soil ecology, and cause soil and water pollution. In this review, we discuss the contribution of F to soil pollution and present certain remedies.

Key Words: Accumulation; Contamination; Dental fluorosis; Environment; Fluorosis; Pollutants; Remediation; Skeletal effects; Toxicity.

INTRODUCTION

The quality of life and the health of the environment are directly related to each other. The increasing incidence of animal and human health problems due to industrial pollution and anthropogenic environmental changes has attracted worldwide attention and efforts to find new remedies to better manage and sustain the environmental component.[1,2] Toxic pollutants may be released to the environment via the air, soil, and water. Stack emissions to the air may add pollutants to the soil which may accumulate in plants via their roots. These pollutants accumulate in the food chain and then affect humans and wild life. Due to variations in natural and anthropogenic activities, global pollution is increasing and leading to contamination of the ecosystem with metals, non-metals, organic compounds, and inorganic compounds. The major contributors to this contamination are pesticides, sewage disposal, insecticides, herbicides, and the uncontrolled discharge of wastes. In the industrialized world, a large part of the population is exposed daily to a variety of chemicals and toxic metals which are harmful for human health. Compounds with the element fluorine are extensively utilized in almost every biological industry, and pollution by the fluoride ion (F) is widespread in the environment. Although F has an anticaries effect when applied topically to the teeth, fluorine is not an essential trace element and is not necessary for the development of healthy teeth and bones.[3,4] Excessive F intake may adversely effect the health of humans, animals, and plants.[5]

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Sources of F: There are many different sources of F but the main sources are the weathering of rocks, industrial emissions, and atmospheric deposition.[6] The most common sources of F are mineral and geochemical stores and a large proportion of the discharge of F into subsoil water takes place through the degradation of rocks containing fluorine.[7] F is among the more abundant elements in earth crust and is present in various rocks with a range of approximately 100–1000 μg/mg, with 625 μg/mg being a typical value. High concentrations of F are present in granites, felsic, quartz monzonites, syenites, biotite, and granodiorites. F-containing rocks such as muscovite, pegmatites, amphibolites, and biotite micas supply F to groundwater and soil by different processes such as soil forming and weathering.[8] There are also anthropogenic sources of F such as the emission of hydrogen fluoride (HF) to the air or the addition of fluoride to water with various human activities, e.g., motorization, fluoridation of drinking water supplies, industrialization, and utilization of F-containing pesticides.[9] Production of phosphate fertilizers is a major industrial source of F. A substantial amount of fluorine, e.g., 3.5%, is present in fertilizer made from rock phosphate but this percentage is reduced in the manufacturing process.[8] Other anthropogenic sources for the entry of F into the earth include the current utilization of chemicals, such as phosphate manures, calcium fluoride (CaF), sodium fluoride (NaF), hydrogen fluoride, sulfur hexafluoride (SF_6), etc. When F is released into the air, it is carried by the wind to the surrounding soil and vegetation and contaminates them. Contamination of soil with F is basically due to utilization of F-containing fertilizers such as phosphorous fertilizers. The F content of soil commonly varies from 150–400 μg/mg and the value may increase in heavy clay soil up to 1000 mg/kg. The contaminated soil ultimately affects human beings through direct contact, the inhalation of vaporized soil, and the use of water contaminated with F by its passage through the soil. The use and production of phosphate fertilizers, the ceramic, zinc and steel industries, and energy plants are noteworthy sources of F pollution in the environment.[10]

Spatial distribution of F: The total concentration of F in the soil is usually derived from the parent material and therefore its distribution pattern in soil is related to the process of soil formation. The average content of F in the soil in world-wide has been estimated as 329 μg/mg. The lowest F content are usually present in sandy soil in relatively humid environments, while the higher concentrations of F are found in soil from weathered mafic rocks and in heavy clay soil. The pH of the soil, clay, and organic carbon content are the prime determinants of soil F content.[11] F enters the soil through different ways such as dry deposition, precipitation, and with contaminated litter where it is absorbed readily. The absorbed F increases the total soluble F concentration in the soil, influences the pH of soil, and can combine with toxic elements such as aluminum and heavy metals. F can exist as the free fluoride ion (F^-) or form complexes with elements such as iron (Fe), boron (B), calcium (Ca), and aluminium (Al), with the complexes of Al and F being most prevalent.[12]

Effects of F pollution: The aerial F emitted from industries not only pollutes the air but also, along with ground water contaminated by fluorine-containing mineral deposits, contaminates plants, crops, soil, vegetables, and freshwater bodies. The use these F-contaminated products and sources may result in toxic effects on health of both humans and domestic animals.[13] While the topical use of F on teeth is regarded as having beneficial effects, the evidence for any beneficial effects from systemic absorption is now considered to be weak.[4]

Impact of F on the human system: The research has shown repeatedly that the consumption of F can extremely be harmful and in some cases deadly. In terms of acute toxicity, F is slightly less toxic than arsenic and more toxic that lead. High concentrations of F can lead to serious poisoning incidents with death.

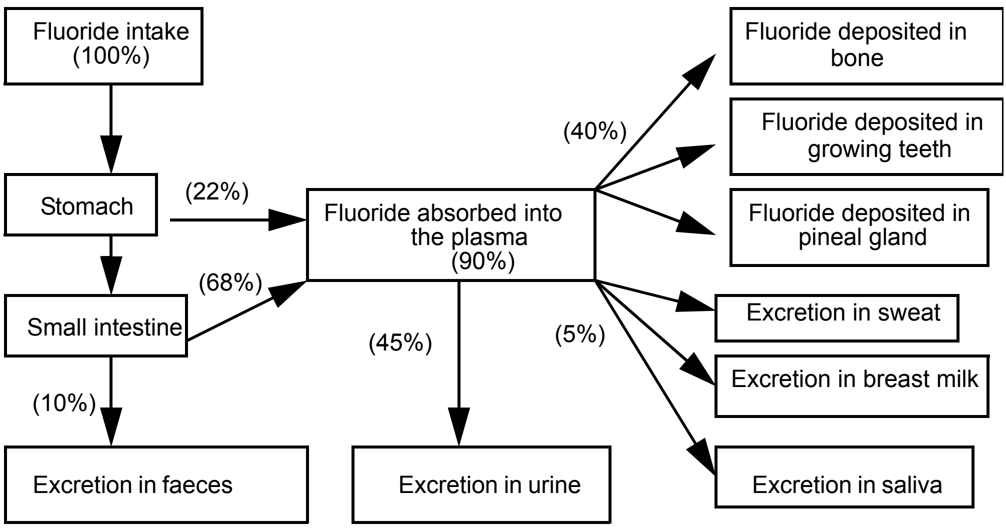


Figure 1. Metabolism of F in the human body.[13-16]

Chronic toxicity can occur with long term exposure. Absorption from the gastrointestinal tract and lungs occurs rapidly with the peak in the plasma occurring after 30 min. Fluoride is cleared from plasma through uptake by bones and excretion in the urine with infants and children clearing relatively larger amounts of F into bones because of their growing skeleton.[14] After a single dose, plasma concentrations rise to a peak and then fall as the F is cleared by the renal system and bone back to a short-term baseline with a half-life of several hours.[14] For each mg of F that is ingested approximately 0.9 mg is absorbed in the stomach and small intestine and passes to the blood while 0.1 mg is excreted unabsorbed in faeces, 0.40 mg is stored in bones and teeth, 0.45 mg is excreted in the urine, and 0.05 mg is excreted in breast milk, sweat, and saliva.[14,15] In the rat, approximately 25% of F absorption occurred in the stomach and 75% from the small intestine.[16] The skeleton and teeth are the prime organs of F retention/accumulation in human body, while relatively small amounts may be deposited in another calcifying organ, the pineal gland.[14,17] The retention period of F can be up to several years. F can reach the foetus via the placenta.[9] (Figure 1).

Fluorosis: A high consumption of F may cause chronic fluorine toxicity, known as fluorosis[1]. One of the mechanisms involved in the pathogenesis of fluorosis is increased oxygen radical generation and lipid peroxidation. Dental fluorosis, with mottling of the teeth, is common in F endemic areas. The teeth develop chalky white patches and become rough. In addition, yellow to dark brown lines may become visible.[17] Yellow to brown, or even become black, opaque patches may occur with more severe toxicity with increased tooth porosity which leads to chipping or pitting.[18] In skeletal fluorosis the mass and density of bone increases along with the development of skeletal and joint symptoms. Severe pain in the spine, joints, and hip area are early symptoms. The symptoms of "poker back" may appear with increasing stiffness until whole spine converts into one continuous column of bone. A number of muscles of the spine may also become calcified and paralyzed as the condition increases in severity. Neurological defects, complete paralysis, crippling deformities of the spine and the major joints, muscle wasting, and increased density of the spinal cord may occur in the most advanced stages of skeletal fluorosis. Skeletal fluorosis may occur with a high F intake from water and other dietary factors.[19]

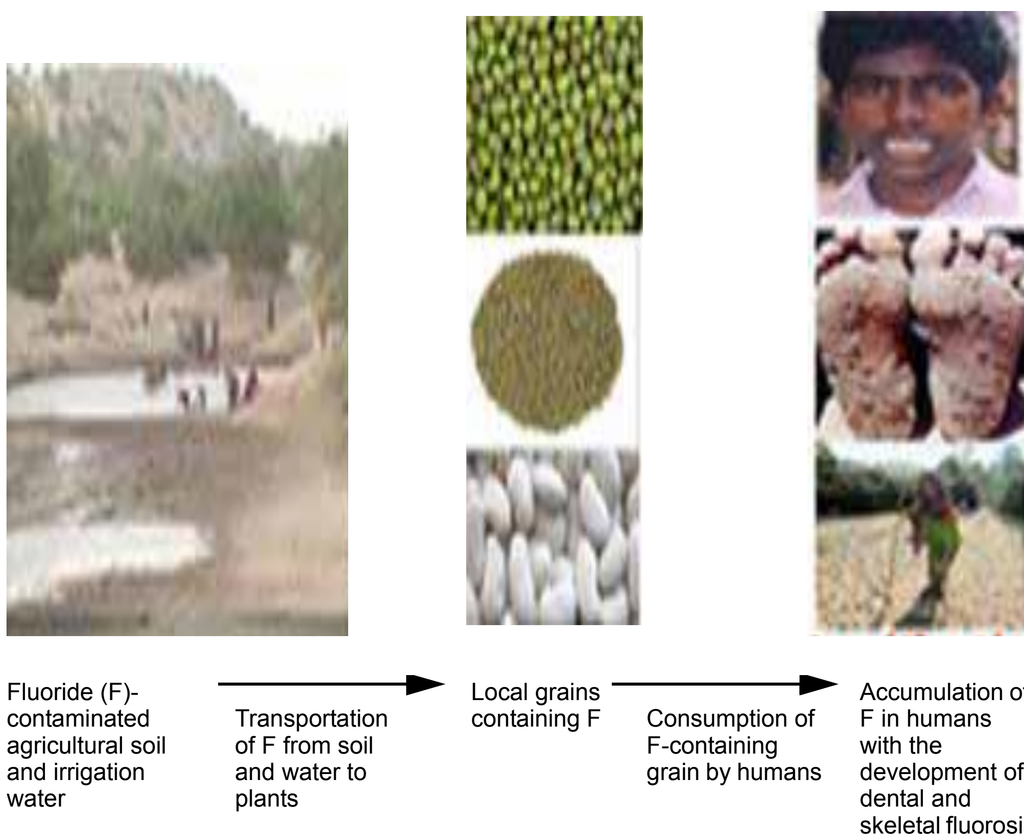


Figure 2: The transportation of fluoride from agricultural soil and irrigation water to plants, the consumption of grains containing fluoride by humans, and the accumulation of fluoride in humans with the development of dental and skeletal fluorosis.[1]

Phytotoxicity of F: Fluorine exists in the environment as gaseous molecules (F_2) and in its reduced form as the ion fluoride (F). Under certain circumstances of comparatively lower pH and hardness fluorine occurs as the fluoride ion in water. Plants are exposed to F through different sources such as air, water, and soil. Natural sources of F include cryolite, apatitite, feldspar, volcanic gases, and marine aerosols.

Other important sources of F include the production of bricks, glass, aluminium, ceramics, and high phosphate fertilizers. The trace amounts of F available to plants by diffusion in the soil may absorbed by roots. In plants such as tea, F is naturally accumulated. In some cases, F unfavorably affects the plant leaves after it intrudes into the leaves because of its high solubility. F deposition may become relatively slower over time. Fluoride may stop photosynthesis and other essential processes in plants. F passes from roots to leaves through the process of transpiration or moves through stomata and accumulates in the margins of the leaves. Marginal and tip necrosis is the first symptom of fluorine injury in plant leaves. However, such symptoms may also appear in drought or salinity stress, which can resemble injury caused by F. Unfortunately, an enormous number of plants are highly F sensitive such as dracaena, Tahitian Bridal Veil, the spider plant, etc. Economically important fruits are also sensitive to F toxicity such as apricot, peaches, plums, etc. Flowers may also be affected by F toxicity, e.g., tulip, gladiolus, lily, etc.[20] Arora and Bhateja reported that F in soil, crops such as wheat, rice, and potato, and in other dietary sources can lead to the occurrence of fluorosis. Therefore, one should carefully calculate the amount of F in daily diet.[21]

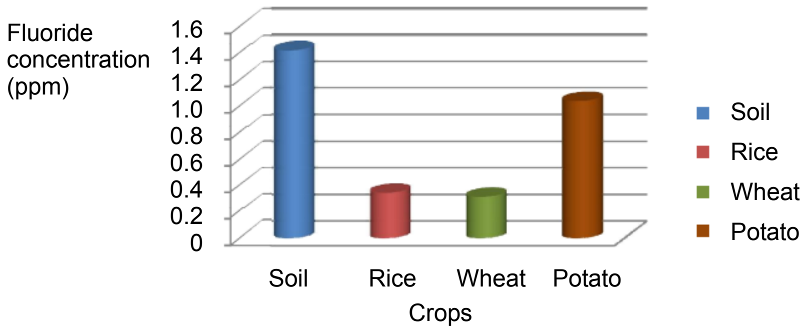


Figure 3. Mean F concentrations (ppm) of different crops in relation to the mean F concentration in the soil.[21]

Mishra at al. reported that accumulation of F in plant tissues is directly proportional to the amount present in soil.[22] A number of these plants were examined and found to have a substantially decreased Net Primary Productivity (NPP) [above ground biomass (AGP) and below ground biomass (BGP)] and yield (pod weight) with exposure to F (Figure 4).

When treated with F in a concentration of 20–100 ppm, the NPP decreased in brinjal, mung, and tomato plants by 6.64–56.72%, 10.27–53.61%, and 14.46–62.24%, respectively, When treated with F in a concentration of 10–50 ppm, mustard, okra, and chillies also showed considerable declines in NPP of 15.58–61.21%, 12.28–52.78%, and 40.8–90.65%, respectively. When treated with F in a concentration of 20–100 ppm, the monocots maize and rice showed a decline in the NPP of 12.17–61.20% and 6.64–56.72%, respectively. Hence, it has been found that F toxicity in soil may harshly effect the NPP in several crops in minute as well as in extreme concentrations.[22]

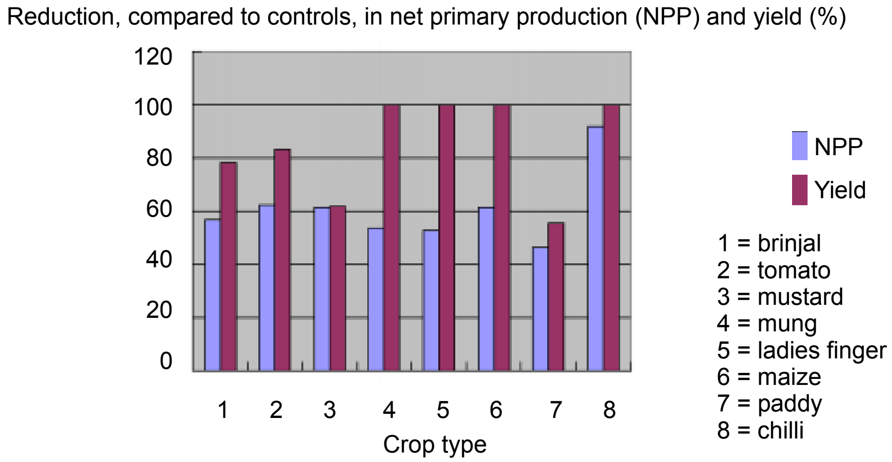


Figure 4. Percent reduction, compared to controls treated with tap water, in net primary production (NPP) [above ground biomass (AGP) and below ground biomass (BGP)] and yield (pod weight) in different crops treated with various concentrations of fluoride ion (F). A 100% reduction in yield indicates that no pods were formed.[22]

1 = brinjal (F concentrations used in the F-treated groups: NPP=100 ppm, yield=100 ppm)
2 = tomato (F concentrations used in the F-treated groups: NPP=100 ppm, yield=1000 ppm)
3 = mustard (F concentrations used in the F-treated groups: NPP=50 ppm, yield=50 ppm)
4 = mung (F concentrations used in the F-treated groups: NPP=100 ppm, yield=100 ppm)
5 = ladies finger (F concentrations used in the F-treated groups: NPP=50 ppm, yield=50 ppm)
6 = maize (F concentrations used in the F-treated groups: NPP=100 ppm, yield=20 ppm)
7 = paddy (F concentrations used in the F-treated groups: NPP=100 ppm, yield=100 ppm)
8 = chilli (F concentrations used in the F-treated groups: NPP=100 ppm, yield=20 ppm)

Due to the high capacity of some plants for the uptake of F from soil the monitoring of soil F levels is necessary from time to time. Spinach is a popular vegetable and has a high capacity for taking up F intake so that this plant may have high F levels and consequently there may be an increased risk of health consequences after its consumption. Therefore, it is required that this type of vegetable be cultivated away from soil that has accumulated high F levels.[23] The accumulation and uptake of F in the shoot and root of spinach (*Spinacea oleracea*) and the effects of contamination of soil by inorganic F (NaF) were investigated in a pot experiment under controlled conditions.[23] Using soil with a soluble F range of 2.57–16.44 mg/kg soil, the experimental results showed that most of the F accumulated in the spinach tissues and the plant had a mechanism for partitioning the water labile F and the total F in its tissues (Figure 5). It restricted the translocation of F from the root to the shoot, thus sparing the part of the plant mostly used for consumption.[23]

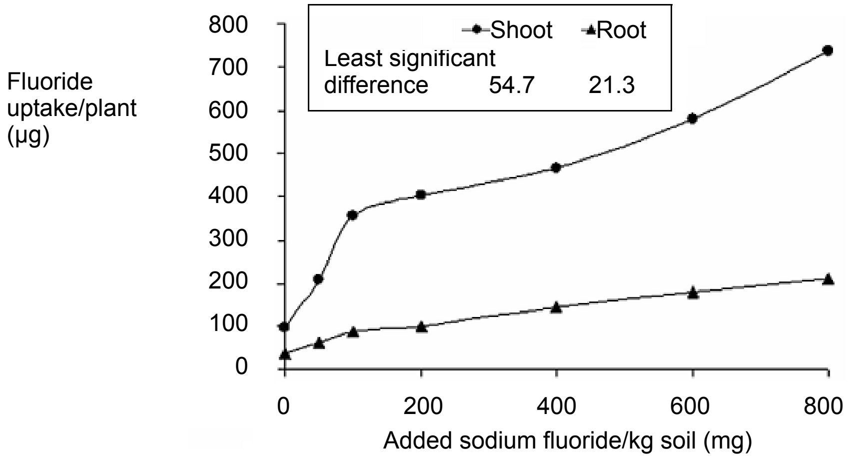


Figure 5. Effect of NaF on the F uptake of *Spinacea oleracea*. LSD (0.05): Least significanc difference between the means at the 0.05 probability level: shoot 54.7, root 21.3.[23]

Several plants have the capability to accumulate soil F through several mechanisms. The potato plant has a high capacity to accumulate soil F and the strong fluorine accumulation in the shoots and leaves indicates the potential of *S. tuberosum* for field application for the removal of fluorine. In the case of the potato, the tolerance index and growth ratio have different trends from each other as concentration of F changes. The F accumulation is highest in the leaves while percentage total F translocation from soil to plant linearly decreases as the soil F increases. Therefore, the potato (*S. tuberosum*) can be a suitable candidate species for the removal of F for phytoremediation purposes.[24]

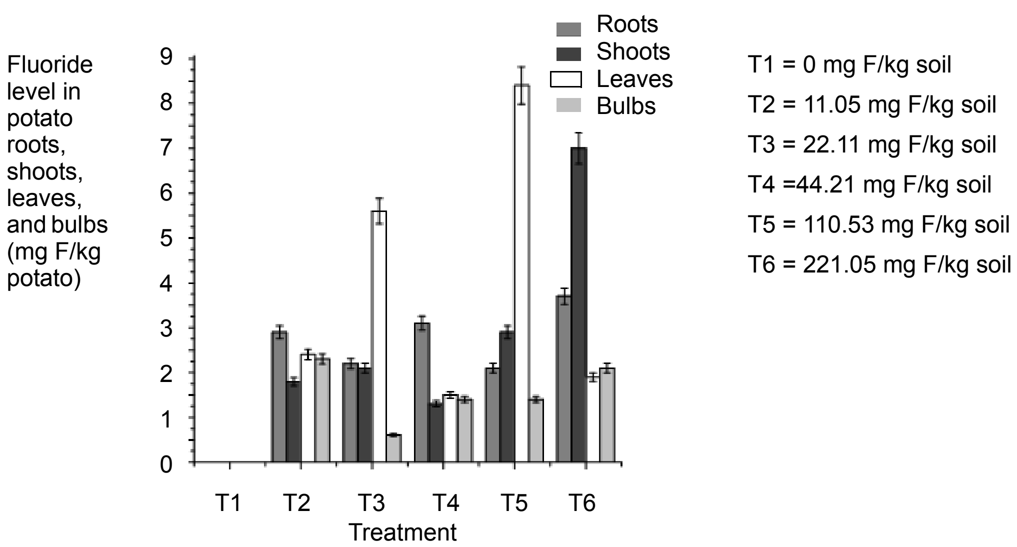


Figure 6. Fluoride accumulation (mg F/kg potato) in potato roots, shoots, leaves, and bulbs at different levels of soil fluoride. The bars represent the least significant difference (LSD) between the means at the 0.05 probability difference.[24]

Tea plants have a high ability to absorb F from soil and surrounding air and accumulate in the leaves in the form of an Al and F complex. Consumption of high F tea for long time can result in chronic F intoxication.[25]

Toxicity of F in birds and animals: The fluoride ion (F) is not considered to be essential for human growth and development,[4] including for the development of healthy teeth and bones, and the chronic ingestion of fluoride at high levels (above 6 mg/day) can be toxic to animals and human beings and cause dental, skeletal, and non-skeletal fluorosis.[26] Fluorosis usually occurs in two forms: (i) endemic fluorosis caused by drinking water or consuming food with a high F content and (ii) industrial fluorosis resulting from exposure to air containing a considerable F content. Fluorosis, in humans, animals, and birds, affects not only the skeletal parts of the body but also affects soft tissues, e.g., brain, liver, kidney, thyroid, and spinal cord. Anjum et al. investigated the effect of a high concentration of F on hepatic and renal enzymes in four groups of domestic chickens, A, B, C, and D, receiving 0, 10, 20, and 30 µg/g of NaF by body weight, respectively, on a weekly basis for four weeks.[27] Alkaline phosphatase (ALP), aspartate aminotransferase (AST), alanine aminotransferase (ALT), and bilirubin were determined as indicators of liver function, while uric acid was used as a parameter for renal function. The results showed high values for all the parameters ($p<0.05$) in the F-treated groups indicating that F effected hepatic and renal function in the exposed birds.[27]

Although all animals are susceptible to high doses of F, the tolerance level changes from one species to another. Noteworthy sources of F for terrestrial animals are drinking water, soil, and vegetation contaminated with F emitted by different activities such as volcanic eruptions and industrial activities[28] (Figure 7). The metabolism of F in animals is similar to that of humans[28] (Figure 8).

Among the terrestrial vertebrates, herbivores are more susceptible than carnivores and other animals. Domestic and wild herbivores are more exposed to environmental F contamination because they are nonselective eaters and can consume contaminated feed, water, and forage. Cattle and sheep have attracted more attention from researchers worldwide, perhaps due to their large populations and their greater economic importance. However, other animals, including water buffaloes, horses, goats, pigs, and wild cervids, can also suffer from F-toxicity naturally.[28] Skeletal, non-skeletal, and dental fluorosis have been studied in buffaloes (*Bubalus bubalis*), camels (*Camelus dromedarious*), donkeys (*Equus asinus*), horses (*Equus caballus*), and cattle (*Bos taurus*).[29]

Choubisa et al. studied F toxicity in 760 domestic animals comprising 386 cattle (*Bos taurus*), 158 goats (*Capra hirus*), 7 donkeys (*Equus asinus*), 11 horses (*Equus caballus*), 131 buffaloes (*Bubalus bubalis*), and 67 sheep (*Ovis aries*) from different villages with a mean water F range of 1.5–4.4 ppm.[30] Three hundred and seventy (48.7%), were found to be afflicted with dental mottling. On clinical examination, 325 (42.8%) of the animals revealed periosteal exostoses, intermittent lameness, hoof deformities, stiffness in the legs and tendons, and wasting of the main mass of the hind quarter and shoulder muscles. Fluorosed animals also showed signs of non-skeletal fluorosis such as gastro-intestinal discomforts, impaired reproductive functions, neurological disorders, and congenital abnormalities. The prevalence and severity of these F effects increased with an increasing F concentration in the water

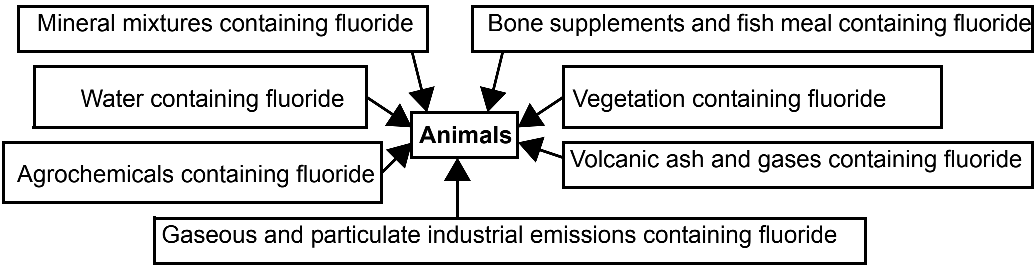


Figure 7. Sources of fluoride in animals.[28]

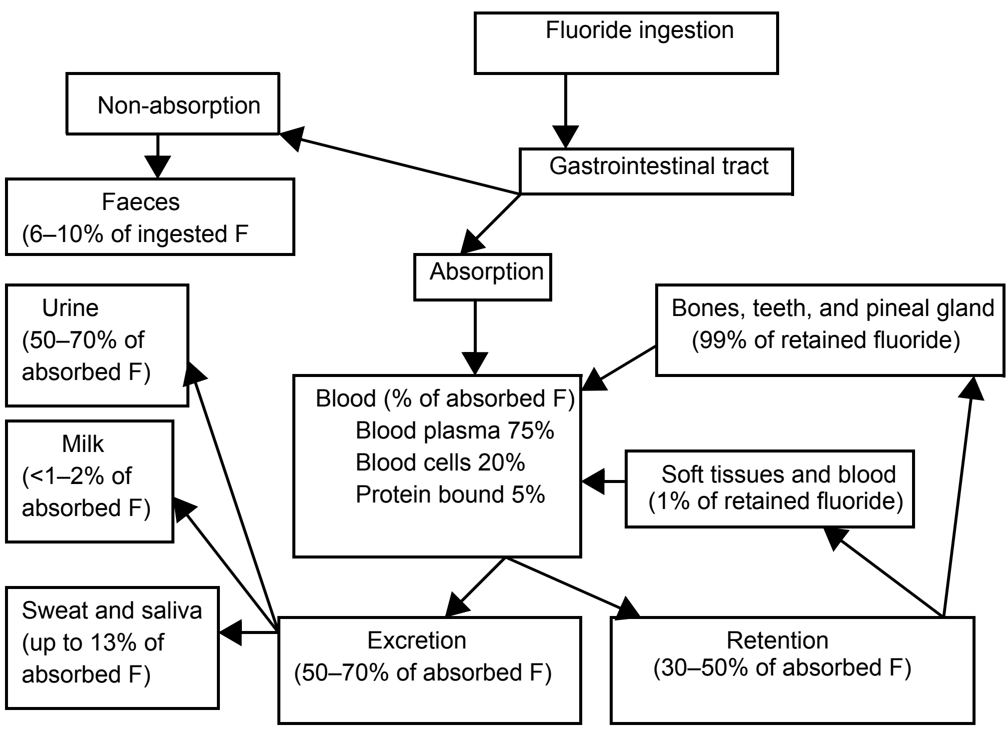


Figure 8. Fluoride metabolism in the animal body after its oral ingestion.[28]

Remediation of F-contaminated soils: Moon et al. described a method for the removal of F from contaminated soil by washing with different solutions such tartaric acid ($C_4H_6O_6$), nitric acid (HNO_3), sulfuric acid (H_2SO_4), sodium hydroxide (NaOH), and hydrochloric acid (HCl) (Figures 9–11).[31] The concentrations of the washing solutions ranged from 0.1 to 3 M with a liquid to solid ratio of 10. The results of washing with the various solutions indicated that HCl was most the effective for removing F from contaminated soils. The highest F removal rate from the contaminated soil, of approximately 97%, was obtained using 3M HCl. The F-removal efficiencies of the washing solutions were in the following order: $HCl>HNO_3>H_2SO_4>NaOH>C_4H_6O_6$.[31]

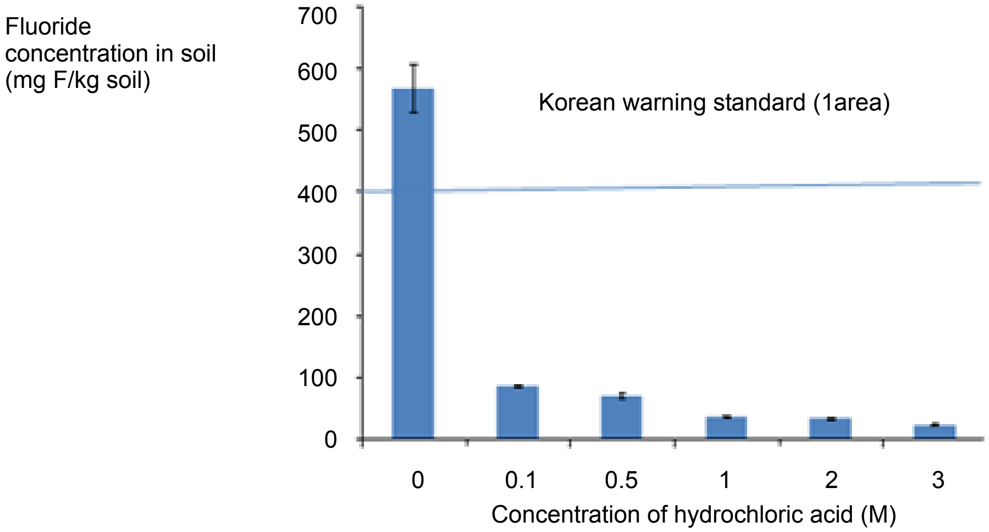


Figure 9. Fluoride concentrations remaining in the soil after hydrochloric acid (HCl) washing at various concentrations together with the Korean warning standard for one area.[31]

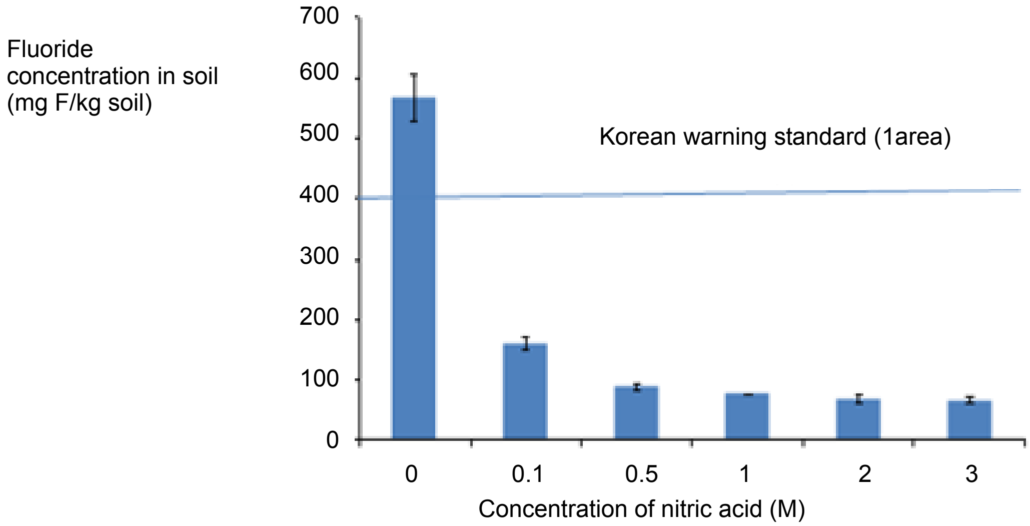


Figure 10. Fluoride concentrations remaining in the soil after nitric acid (HNO_3) washing at various concentrations together with the Korean warning standard for one area.[31]

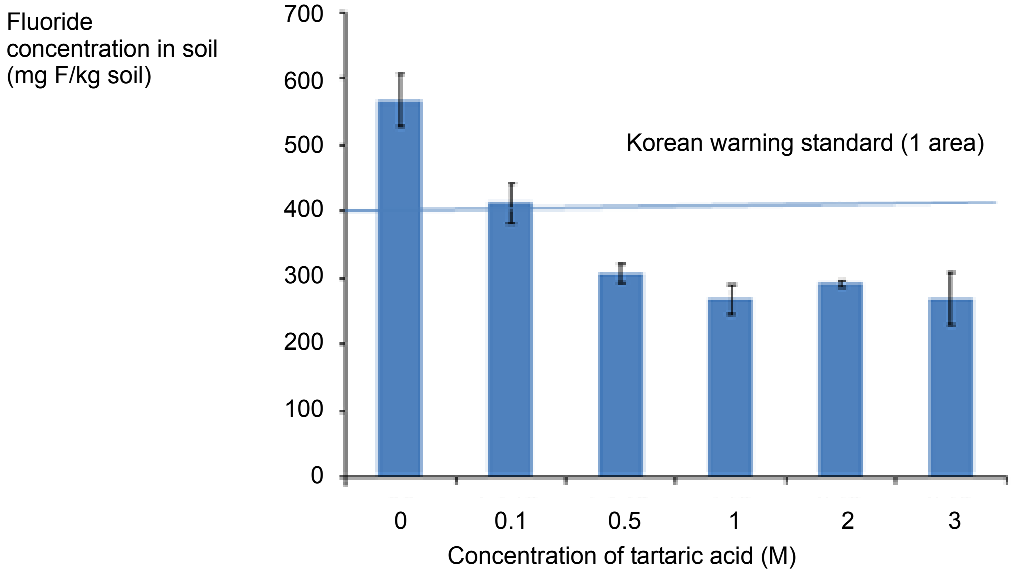


Figure 11. Fluoride concentrations remaining in the soil after tartaric acid ($C_4H_6O_6$) washing at various concentrations together with the Korean warning standard for one area.[31]

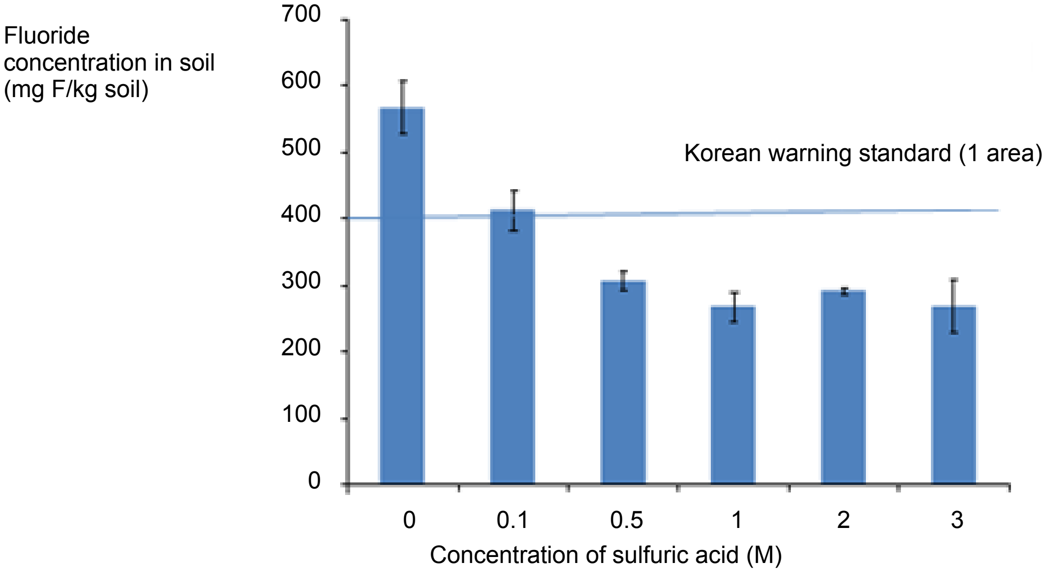
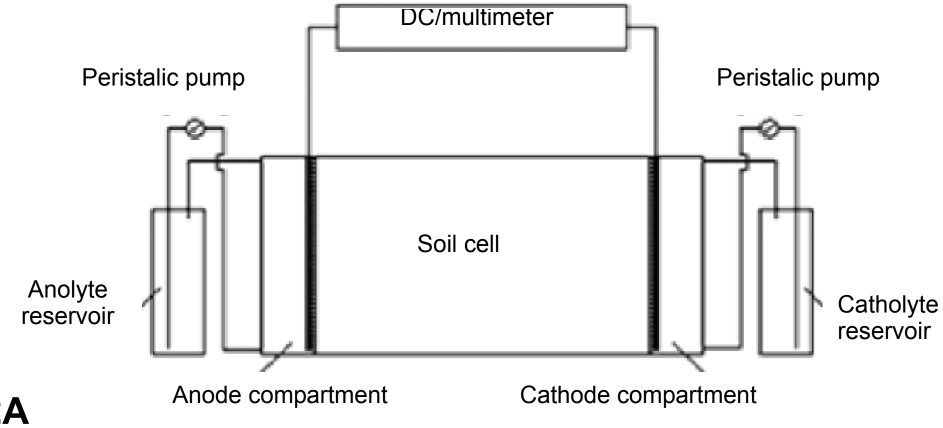
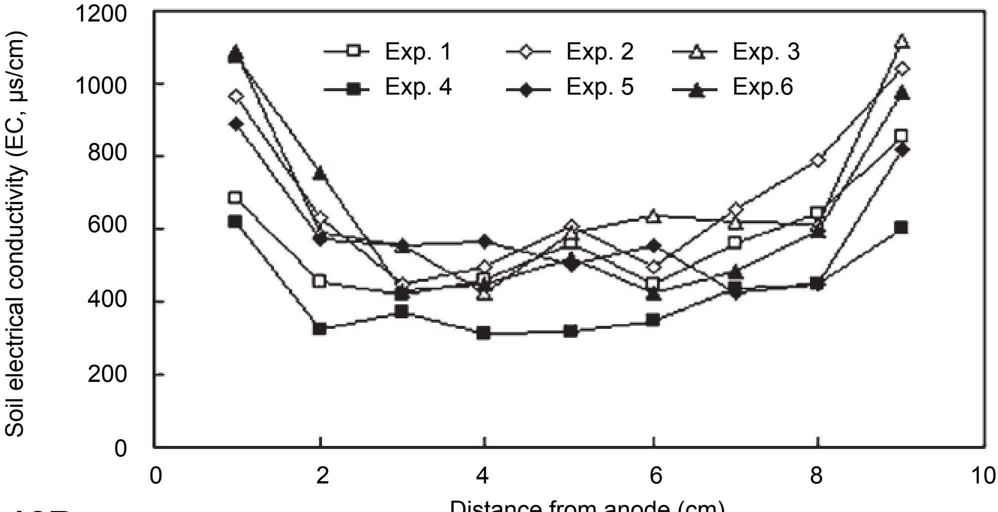


Figure 12. Fluoride concentrations remaining in the soil after sulfuric acid (H_2SO_4) washing at various concentrations together with the Korean warning standard for one area.[31]

Zhu et al. investigated the electrokinetic remediation of F-contaminated field soil with an organic content, pH, and initial F-concentration of 20.52 g/kg, 18.17, and 1058 mg/kg, respectively.[32] Electrokinetic experiments were carried out under two different concentrations of alkaline solution and three different voltage gradients (Figures 12A-12D). The removal efficiency of fluorine increased up to 73% within 10 days with increasing the concentration of the alkaline solution and the applied voltage. This process could effectively promote the migration of F present in soil. The main transport mechanism was electromigration and the electroosmotic flow had an effect on the soil F migration. An appropriate anolyte enhanced electrokinetic method can be applied to remove fluorine from contaminated field soil and also has a significant potential for removing other anionic pollutants such as arsenic and chromium from soil.[32]

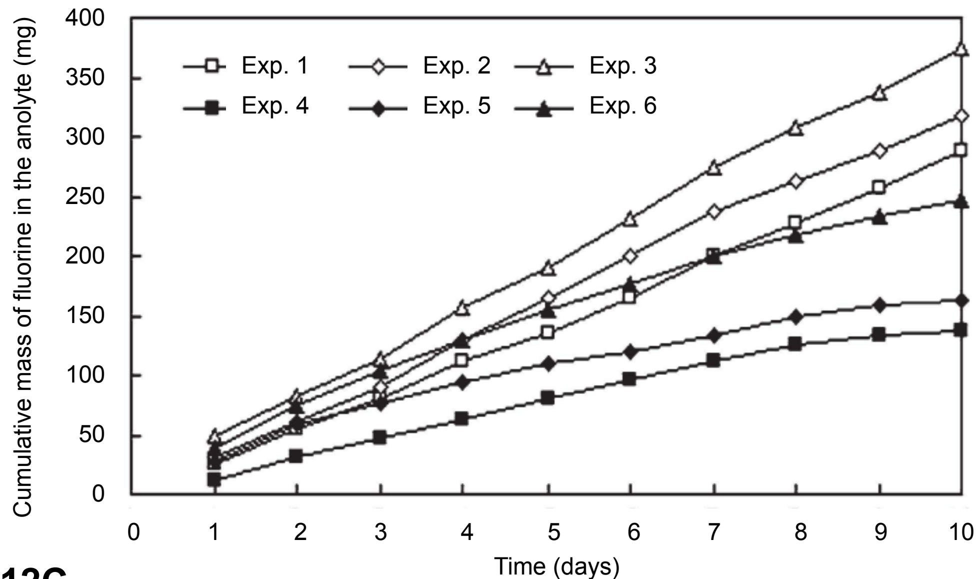


12A

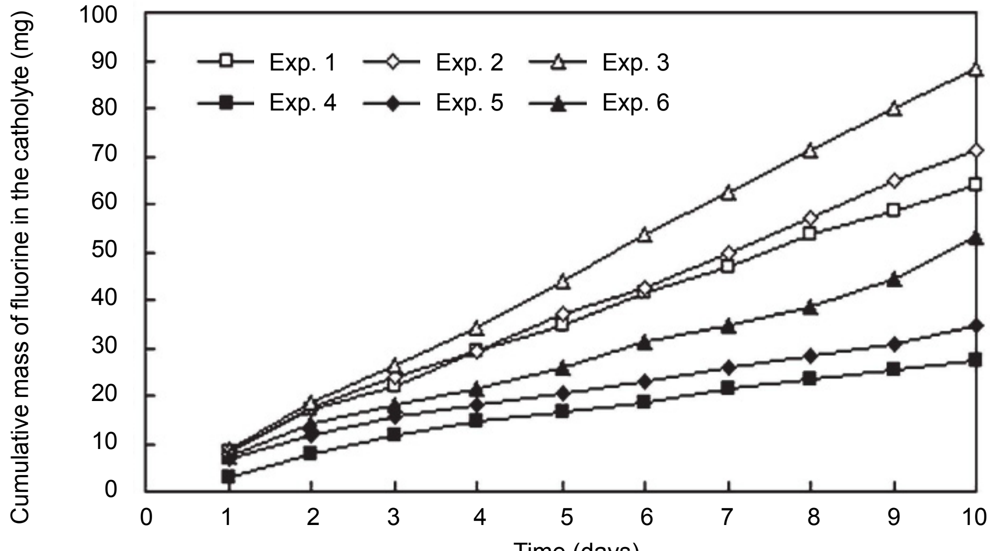


12B

Figures 12A and 12B. 12A: Schematic diagram of the electrokinetic apparatus; 12B: Soil electrical conductivity (EC, µs/cm) in soil sections after the electrokinetic treatments.[32]



12C

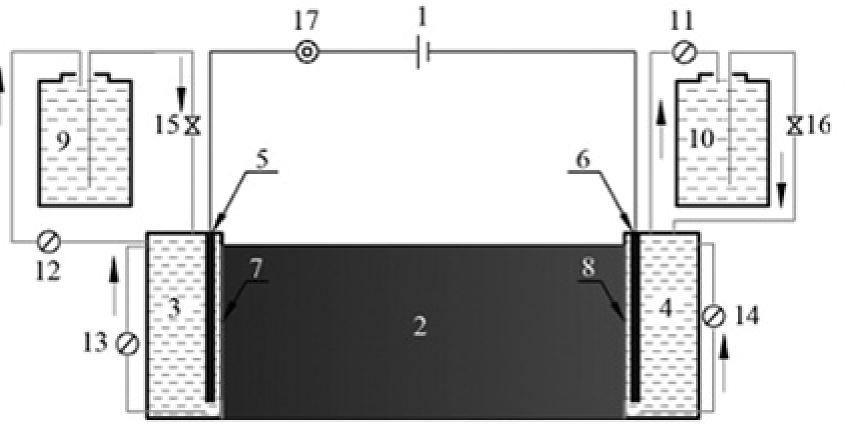


12D

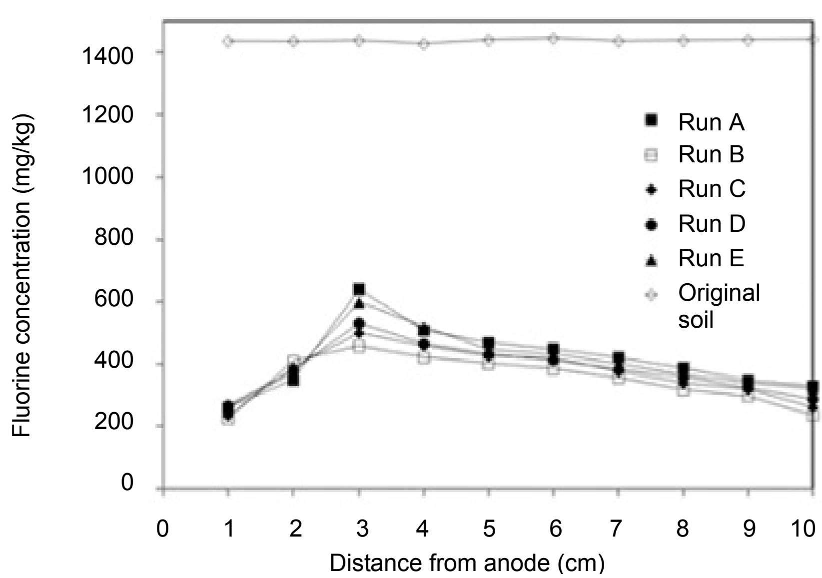
Figures 12C and 12D. 12C: Cumulative mass of fluorine in the anolyte (mg); 12D: Cumulative mass of fluorine in the catholyte (mg).[32]

Zhou et al. reported the effect of changing the pulse interval of an electric field on the remediation of fluorine-contaminated soil by an electrokinetic remediation method.[33] The experiment results indicated that at the same intensity of the electric field, pulse-enhanced electrokinetic remediation showed a better performance in the removal of fluorine than conventional electrokinetic remediation. The efficiency of the pulsed enhanced electrokinetic remediation was increased because concentration polarization decreased with applying the electric field and it increased the electric current, the electrical voltage, and the electroosmotic flow in the soil cell. Therefore, for the removal of fluorine from soils, a pulse-enhanced technique would be a preferred choice.[33]

13A

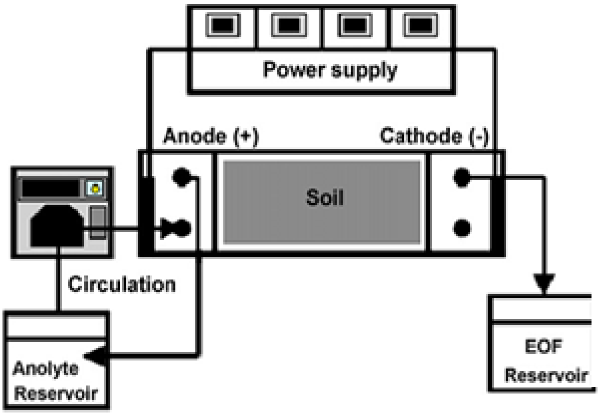


13B



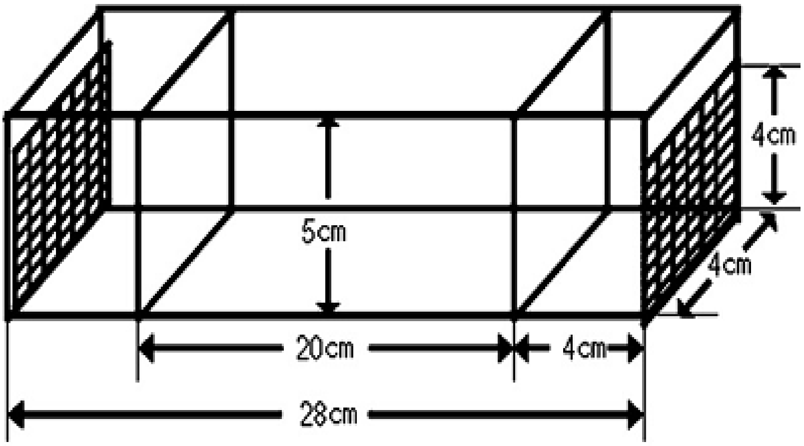
Figures 13A and 13B. 13A: A schematic diagram of the experimental apparatus. Key to numbering: 1. The DC power supply (GPC6030D, Gw instek, China); 2. The soil cell (10 [L] cm×6 [W] cm×8 [H] cm); 3, 4. The electrode compartment (4 [L] cm×6 [W] cm×8 [H] cm); 5, 6. Working electrode (graphite sheet, 1 [L] cm×6 [W] cm×8 [H] cm); 7. Anion exchange membrane (3361BW, Shanghai Shanghua Water Treatment Material Co., Ltd.); 8. Cation exchange membrane (3362BW, Shanghai Shanghua Water Treatment Material Co., Ltd.); 9, 10. The electrolyte reservoir; 11-14. 4-Channel peristaltic pump (BT00-300T/DG-4, longerpump, China); 15, 16. Simple flow regulator; 17. Time; 13B: Residual fluorine concentration in soil sections after electrokinetic remediation.[33]

Kim et al. studied electrokinetic remediation for the removal of fluorine from contaminated soil and the results showed that electro kinetic techniques increased the efficiency of fluorine removal by up to 75.6%.[34] From these results it can be concluded that analyte conditioning is a very effective enhancement method to remove fluorine from contaminated soil in electrokinetic remediation. This method can be also applied for removal of anionic pollutants such as chromate and arsenic from contaminated soil.



14A

EOF = electroosmotic flow



14B

Figures 14A and 14B. Schematic diagram and dimensions of the experimental apparatus: (A) anolyte circulation system; (B) dimensions of the experimental apparatus.[34]

CONCLUSIONS

This review considers the importance of F and argues that a high intake of F, via ingestion or inhalation from a variety of sources, may cause toxicity in humans and animals, including dental, skeletal, and non-skeletal fluorosis. The F toxicity can be acute or chronic depending on the level and the duration of the F-intake. The studies in the literature show that the presence of a high concentration of F in soil affects plants and aquatic life and leads to soil and water pollution. Plants species with a susceptibility to F pollution in soil may be drastically damaged. In addition, F pollution may have a devastating effect on the microbial activity in soil and disrupt the soil ecology.

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