REVIEW ARTICLE



# Status of industrial fluoride pollution and its diverse adverse health effects in man and domestic animals in India

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Abstract Hydrofluorosis in humans and domestic animals is a worldwide health problem and caused by a prolonged period of fluoride exposure through drinking of fluoride contaminated water. But in recent years, due to rapid industrialization in India, diverse serious health problems among industrial workers and residents and domestic animals living in the industrial areas due to fluoride pollution are on the rise. A number of coal-burning and industrial activities such as powergenerating stations, welding operations and the manufacturing or production of steel, iron, aluminum, zinc, phosphorus, chemical fertilizers, bricks, glass, plastic, cement, and hydrofluoric acid are generally discharging fluoride in both gaseous and particulate/dust forms into surrounding environments which create a industrial fluoride pollution and are an important cause of occupational exposure to fluoride in several countries including India. An industrial emitted fluoride contaminates not only surrounding soil, air, and water but also vegetation, crops and many other biotic communities on which man and animals are generally dependants for food. Long- time of inhalation or ingestion of industrial fluoride also

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causes serious health problems in the forms of industrial and neighborhood fluorosis. In India, whatever research works conducted so far on the chronic industrial fluoride intoxication or poisoning (industrial and neighborhood fluorosis) in man and various species of domestic animals due to a prolonged period of industrial fluoride exposure or pollution (contamination) are critically reviewed in the present communication. Simultaneously, we are also focused the various bioindicators and bio-markers for chronic industrial fluoride intoxication or pollution.

Keywords Bio-indicators . Bio-markers . Contamination . Animals . Hydrofluorosis . Industrial fluoride . Industrial fluorosis . Neighborhood fluorosis . Osteo-dental fluorosis . Pollution . Residents . India

# Significance and source

## Fluorine/fluoride

Fluorine (F<sup>−</sup> ), the ninth element of the periodic table was discovered by Professor Henry Moissan on June 26, 1886. However, its applications and biological significances came to be known very late in the decade of 1920. Owing to its electronic configuration  $(1s^2, 2s^2, sp^5)$ , fluorine is the most electronegative and highly reactive diatomic pale yellowgreen with a pungent and irritable gas; hence, not found free in the nature in element form. It has strong affinity to combine chemically with other elements to form compounds called fluorides (F). Many researchers and reviewers use the word fluoride to denote the ionized, physiologically available form of the element. The word *fluorine* has sometimes been thought to denote exclusively the gaseous form of the element. However, by analogy with iodine, fluorine may be used to

denote collectively the element in all its forms ionized or ionisable. Where, there is doubt as regards to ionizability, e.g., in certain foods, fluorine is actually the better, more comprehensive word (WHO [1970](#page-10-0)).

### Sources of fluorine/fluoride

Fluorine is seventeenth in the order of abundance in the earth crust (WHO [1970](#page-10-0)) and is varyingly distributed in sea water, drinking water, soil, dust, and in mineral deposits of sallaite (MgF<sub>2</sub>), villiaumite (NF), fluorspar (CaF<sub>2</sub>), cryolite (Na<sub>3</sub> Al  $F_6$ ), bastnaesite (CeLaY) (CO<sub>3</sub>)F, fluoroapatite  $\lceil Ca_5(PO_4)_3 \rceil$ etc., with concentrations (%) of 61, 55, 49, 45, 9, and 3.5, respectively. Fluoride concentration in the atmosphere in unpolluted areas usually varies between 0.02 and 2.0  $\text{prg/m}^3$ (USEPA [1980](#page-10-0)). Atmospheric fluoride may be in gaseous or particulate forms. Most common gaseous forms include hydrogen fluoride (HF), sulfur hexafluoride (SF $_6$ ), silicon tetrafluoride (SiF<sub>4</sub>), hexafluorosilic acid (H<sub>2</sub>SiF<sub>6</sub>), and carbontetrafluoride  $(CF_4)$ . Particulate forms include sodium aluminum fluoride (NaAlF<sub>3</sub>), calcium phosphate fluoride (CaFO<sub>4</sub>P), sodium hexa fluorosilicate (F<sub>6</sub>Na<sub>2</sub>Si), aluminum fluoride (AlF<sub>2</sub>), calcium fluoride (CaF<sub>2</sub>), and lead fluoride  $(PbF<sub>2</sub>)$ . Hydrogen fluoride and inorganic fluoride particulates (sodium and calcium fluoride) are major inorganic fluorides present in the atmosphere, accounting for nearly 75 and 25 %, respectively (WHO [1970\)](#page-10-0).

The principal sources of fluoride to man and animals are: drinking water, vegetation grown on fluorotic soils and water, certain edible marine animals, fluoride rich phosphate feed supplements, mineral mixture, medicines, cosmetics, dust in air, and certain industrial processes (Swarup and Dwivedi [2002;](#page-10-0) Ranjan and Ranjan [2015\)](#page-10-0). However, in India, the groundwater for drinking is the main source of chronic fluoride intoxication or poisoning in both man and domestic animals. In some areas, bore well water contained a much higher fluoride content compared to surface water (Underwood and Suttie [1999\)](#page-10-0).

In India, chronic fluoride poisoning is endemic in several states due to excess quantity of fluoride in drinking water sources, especially in the rural areas where this health problem is relatively more prevalent (Choubisa et al. [1995,](#page-9-0) [1996a,](#page-9-0) [2001;](#page-9-0) Choubisa [1997\)](#page-9-0). Among the states in India, 50–100 % districts are affected with fluoride intoxication in Andhra Pradesh, Gujarat, Rajasthan and Uttar Pradesh. While in Bihar, Haryana, Karnataka, Madhya Pradesh, Maharashtra, Punjab and Tamil Nadu and 30–50 % districts are endemic for fluoride intoxication (Choubisa [2012a](#page-9-0)).

For the purpose of economy growth and sustainable development, industrialization is essential. But due to rapid industrialization in India, side by side is also creating many health problems not only in the people but also in domestic animals living in the surrounding areas of several fluoride discharging industries. These are coal-burning and industrial activities such as power generating stations, welding operations and the manufacturing or production of steel, iron, aluminum, zinc, phosphorus, chemical fertilizers, bricks, glass, plastic, cement, and hydrofluoric acid that generally release fluoride in both gaseous and particulate/dust forms into surrounding environments (WHO [1970;](#page-10-0) Swarup and Dwivedi [2002](#page-10-0)). Ultimately, the emitted industrial fluoride deposits on the ground and the herbage/vegetation. The principal hazard of ingesting fluoride contaminated herbage for a prolonged period is the development of animal fluorosis. The chronic inhalation or exposure of fluoride from such polluted or contaminated environment causes dental, skeletal and non-skeletal disorders or adverse changes in industrial workers/ employees as well as in residents living in the vicinity of such industries (WHO [1970\)](#page-10-0).

Fluorine occurred naturally in rock, particularly in association with phosphate. The soils derived from rocks and surface water leaching through the rocks might contain toxic quantities of fluoride (Radostits et al. [1994\)](#page-10-0). The supplementation of rock phosphate and fertilizer grade phosphorous supplement (monoammonium and diammonium phosphate) may cause chronic fluoride toxicity in animals (Jubb et al. [1993\)](#page-10-0). The bones of mature human and animals had higher fluoride even in the absence of abnormal exposure to fluoride content. The bone meals could therefore constitute a significant source of fluorine for farm animals (Underwood and Suttie [1999](#page-10-0)).

## Industrial and neighborhood fluorosis

For a prolonged period of fluoride exposure through water, food, soil and air causes diverse adverse health or toxic effects in the form of fluorosis in mammals especially in man and domestic animals. If these effects are generated by drinking of fluoridated water, then they are collectively referred to as hydrofluorosis which is natural, more prevalent and wide spread in nature. In contrast, neighborhood fluorosis and industrial fluorosis are anthropogenic, relatively less prevalent, restricted to a particular location/place or herd, and caused by the long-term exposure to fluoride emitted from the different industrial operations. The term industrial fluorosis is generally used and restricted for the industrial workers/employees and domestic animals, whereas neighborhood fluorosis for people and domestic animals residing around the fluoride emitting industries. However, hydrofluorosis is rampant almost in every state of the country (Choubisa et al. [2001](#page-9-0); Choubisa [2012a\)](#page-9-0).

Industrial fluorosis was described for the first time in the world by Bartolucci ([1912\)](#page-9-0) in cattle on a farm close to a superphosphate factory in Italy. He originally called it *osteitis*, tracing it to fluorine originating from the factory. He used the term fluorosis first in a 1927 article (Bartolucci [1927](#page-9-0)), as a

<span id="page-2-0"></span>synonym to *cachexie fluorique*, like Christiani and Gautier [\(1925\)](#page-10-0) did earlier. In India, industrial fluorosis was recognized for the first time in cattle living around the aluminum factory located in the state of Orissa by Rao and Pal ([1978](#page-10-0)). Farm animals were suffering from lameness and this problem was confirmed by pathological and chemical examination of the bones of affected animals. Enormous values for fluorine were found, 1.0 to 2.7 % expressed on the bone ash, as compared with normal values of order of 0.05 to 0.08 %. Fluoride concentration in urine reflected the current fluoride intake and was highly related to length of exposure or total fluoride consumed. The affected cattle were found to be excreting large amounts of fluorine in the urine, even up to 68 ppm as compared with less than 5 ppm in urine of cattle from other areas. The level of fluoride in the urine was indicative of industrial fluorosis (Vandermissen et al. [1993\)](#page-10-0).

In humans, chronic industrial fluoride intoxication was recognized for the first time in Danish cryolite workers by Moller and Gudjonsson ([1932](#page-10-0)). Following the identification and thorough investigations of several cases of osteo-dental fluorosis due to the inhalation of fluoride dusts by Roholm [\(1937\)](#page-10-0), then industrial fluorosis was reported in human beings (WHO [1970](#page-10-0); Franke et al. [1975](#page-10-0); Carnow and Conibear [1981](#page-9-0); Barriga et al. [1997;](#page-9-0) Zhang and Cao [1996;](#page-10-0) Wu et al. [2004\)](#page-10-0) and domestic animals (Blakemore et al. [1948](#page-9-0); Agate et al. [1949;](#page-9-0) Burns and Allcroft [1964;](#page-9-0) Rosenberger and Grunder [1968](#page-10-0); Shupe [1970](#page-10-0); Riet-Correa et al. [1987;](#page-10-0) Shupe et al. [1992;](#page-10-0) Wang [1987;](#page-10-0) Findaci and Sel [2001\)](#page-10-0) from several countries. But studies on neighborhood fluorosis or industrial fluoride poisoning on people living in the surrounding areas of various industries are still limited (Murray and Wilsion [1946](#page-10-0); Murray [1950](#page-10-0)). However, in India, for the first time, Bhawsar et al. [\(1985a](#page-9-0), [b](#page-9-0)) did excellent survey study on neighborhood fluorosis conducted in the 7059 individuals of 15 villages and three urban residential areas within a radius of 3500 m from the fluoride-processing industry (manufacturing a refrigeration gas, Freon) on the west coast of India, north Bombay.

In India, most of the studies are conducted on hydrofluorosis in the humans (Choubisa et al. [1996b](#page-9-0), [1997](#page-9-0); Choubisa and Verma [1996;](#page-9-0) Choubisa [1998](#page-9-0), [1999a](#page-9-0), [2001,](#page-9-0) [2012b](#page-9-0)) as well as in the diverse species of domestic animals viz., cattle (Bos taurus), buffaloes (Bubalus bubalis), horse (Equus caballus), donkeys (E. asinus), camels (Camelus dromedarius), sheep (Ovis aries) and goats (Capra hircus) (Choubisa et al. [1996c](#page-9-0), [2011a,](#page-10-0) [2012;](#page-10-0) Choubisa [1999b,](#page-9-0) [2007,](#page-9-0) [2008,](#page-9-0) [2010a,](#page-9-0) [b,](#page-9-0) [2013a\)](#page-9-0) belonging to different geographical provinces with varying grade of fluoride in drinking water. Despite having a number of different types of industrial activities in the country, only few scientific reports on neighborhood fluorosis in people living in the vicinity of industrial activities (Desai et al. [1988](#page-10-0), [1993](#page-10-0); Samal and Naik [1988](#page-10-0); Stanley and Pillai [1999;](#page-10-0) Sharma and Pervez [2004;](#page-10-0) Pandey and Pandey [2011;](#page-10-0) Pandey et al. [2014](#page-10-0); Choubisa and Choubisa [2015](#page-9-0)) and industrial fluorosis in industrial workers/employee and animals (Naik and Samal [1985,](#page-10-0) [1986;](#page-10-0) Swarup and Singh [1989;](#page-10-0) Samal and Naik [1992](#page-10-0); Singh



Figs. 1–5 Polymorphism and severity of dental fluorosis. Children showing pitting/patches (Fig. 1), fine dots (Figs. 2 and 3) and bilateral stratified dark-yellowish lines on maxillary central incisors (Fig. 4). Excessive abrasions (corrosions) of enamel and expose of cementum and dentine material with deep-yellowish pigmentation, diastemas between teeth, pronounced loss of tooth supporting bone with recession and bulging of gingiva (Fig. 5) (Choubisa [2012b](#page-9-0))

<span id="page-3-0"></span>and Swarup [1994;](#page-10-0) Swarup et al. [1998](#page-10-0), [2001;](#page-10-0) Patra et al. [2000](#page-10-0); Saho and Ray [2004](#page-10-0); Mishra and Pradhan [2007;](#page-10-0) Susheela et al. [2013;](#page-10-0) Kumar and Aravindaksha [2015;](#page-10-0) Choubisa [2015](#page-9-0)) are available. In the present communication, we have critically and briefly reviewed the work done so far by researchers on chronic industrial fluoride intoxication (neighborhood and industrial fluorosis) in man and domestic animals living in the vicinity of various fluoride emitting factories located in different states of the country. Simultaneously, bio-indicators and bio-markers for chronic fluoride toxicosis are also focused in the present review.

## Industrial fluoride pollution or contamination

Fluoridated mineral deposits generally contaminate groundwater but an industrial emitted fluoride pollutes not only the air or atmosphere but also the plants (forage), crops (fodder), vegetables, soil, and freshwater bodies. These fluoride contaminated sources are potential to develop toxic or diverse adverse health effects (fluorosis) in both man and domestic animals. Industrial fluoride contamination can be evaluated and inferred by the estimation of fluoride content in biological (milk, blood serum, urine, teeth, bones, nails etc.) and environmental (soil, water, forage, fodder etc.) samples collected from the surrounding areas of potential sources of fluoride emission (Bhawsar et al. [1985a,](#page-9-0) [b;](#page-9-0) Mukherjee et al. [2003\)](#page-10-0). The presence of fluoride content in any of these samples confirms the persistence of industrial fluoride pollution in the surrounding environment. In a large epidemiological survey study on neighborhood fluorosis conducted in western India, simultaneously spot urine samples of 395 individuals were also tested for fluoride content (Bhawsar et al. [1985a,](#page-9-0) [b](#page-9-0)). These samples were collected from three distinct distances, 500–1500, 1500–2500, and 2500–3500 m from the fluoride processing industry. The mean fluoride concentration (ppm) in these samples was found as 1.89, 1.93, and 2.14, respectively. Urine sampling was also done from different direction in relation to industry. The content of fluoride was found as 2.18 ppm in NE, 2.12 ppm in NW, 1.54 ppm in SE, and 1.91 ppm in SW. These results indicate that fluoride content in the biological sample varies greatly in relation to

distance and direction from the fluoride releasing industry, but it also indicates the persistence of industrial fluoride contamination or pollution in the surrounding environment. In other study the industrial fluoride intoxication in domestic sheep grazing in the surrounding areas of aluminum factory located in Hirakund town of western Orissa was carried out (Samal and Naik [1992\)](#page-10-0) by estimating fluoride content in the biological (milk, urine, blood serum, etc.) as well as in the environmental (forage or legumes and graminoides) samples by recommended method. The forage samples collected from 16 sites around the aluminum factory revealed fluoride concentration in the range of 24.0–360.0 ppm. Fluoride content in these samples collected near to the production site was also found relatively high. Fluoride content in milk, urine and blood serum samples collected from sheep (1–7 years age) within 3 km radius of the aluminum factory also revealed variable values as 0.30–1.60, 10.2–57.6, and 0.41– 1.32 ppm, respectively. In a recent study, fluoride content in urine, blood serum and nail ashes of 462 smelter workers was evaluated and it also found in variable quantity as 5.09, 0.14, and 1.49 ppm, respectively (Susheela et al. [2013\)](#page-10-0). In many other studies conducted in the surrounding areas of superphosphate fertilizer and aluminum factories in different parts of the country, a variable content of fluoride in biological and environmental samples has also been observed (Swarup et al. [1998;](#page-10-0) Patra et al. [2000](#page-10-0); Saho and Ray [2004](#page-10-0); Pandey and Pandey [2011](#page-10-0); Kumar and Aravindaksha [2015](#page-10-0)). Whatsoever, the presence of fluoride in any of these samples collected from industrial areas indicate the presence of fluoride pollution but it does not infer the endemic osteo-dental fluorosis in man and animals until unless the appearance of dental mottling and/or bone deformities. However, these studies indicate that the test of urine is better than other samples because of it contains relatively a higher amount of fluoride as compared to its counter parts.

## Fluoride induced adverse health effects

Chronic fluoride exposure either through consumption of fluoridated drinking water or fluoride emitted from the factory for a long-time results in its accumulation predominantly in



Figs. 6 and 7 Dental fluorosis in mature sheep (Fig. 6) and goat (Fig. 7) rearing in the hydrofluorosis endemic areas characterized with excessive abrasion and homogenous and vertical deep-brownish staining on enamel surface of incisor teeth [\(Choubisa et al. 2011c\)](#page-10-0)

<span id="page-4-0"></span>Figs. 8 and 9 Dental fluorosis in cattle (Fig. 8) and buffalo (Fig. 9) calves rearing in the hydrofluorosis endemic areas characterized with stratified and horizontal deep-brownish staining of incisor teeth ([Choubisa et al. 2011c](#page-10-0))



hard tissues such as teeth and bones causing diverse adverse changes that appear in the form of dental mottling (dental fluorosis) and bone deformities (skeletal fluorosis) in man (Choubisa and Sompura [1996;](#page-9-0) Choubisa [1998](#page-9-0), [2001](#page-9-0), [2012b,](#page-9-0) [c;](#page-9-0) Choubisa and Choubisa [2015](#page-9-0)) and domestic animals (Choubisa [2000,](#page-9-0) [2007](#page-9-0), [2008,](#page-9-0) [2013b](#page-9-0), [2015;](#page-9-0) Choubisa and Mishra [2013](#page-9-0)). Besides these maladies, non-skeletal fluorosis

Figs. 10–13 Severe dental fluorosis in a mature cow (Fig. 10) and a buffalo (Fig. 11). Moderate dental fluorosis in a camel (Fig. 12) and a goat (Fig. 13) living in areas with 1.5– 1.7 ppm fluoride in their drinking waters [\(Choubisa et al. 2011c\)](#page-10-0)

or toxic effects of chronic fluoride exposure in soft tissues or organs viz., gastrointestinal discomforts, neurological disorders, impaired endocrine and reproductive functions, teratogenic effects, renal effects, genotoxic effects, apoptosis, excitotoxicity, etc., have also been reported in man as well as in domestic and laboratory animals. Recently these effects have been scientifically reviewed (Choubisa [2012a\)](#page-9-0).



#### Dental mottling or dental fluorosis

Dental mottling is the earlier pathognomonic sign of all kinds of chronic fluoride poisoning. In general, it is characterized by the presence of bilateral striated and horizontal light to deepbrownish or dark staining on enamel of teeth in humans (Figs. [1](#page-2-0)–[5](#page-2-0)) as well as in immature (Figs. [6](#page-3-0)–[9](#page-4-0)) and mature (Figs. [10](#page-4-0)–[13\)](#page-4-0) animals (Choubisa [2001,](#page-9-0) [2012a](#page-9-0), [2013a,](#page-9-0) [b](#page-9-0)). In some cases dental fluorosis was also seen in the form of light to deep-brownish spots, patches and fine dots or granules on the enamel surface of the anterior teeth. In the severe form of dental fluorosis, pronounced loss of teeth supporting the alveolar bone with recession and swelling of gingival and excessive abrasion or irregular wearing of teeth (Fig. [5](#page-2-0)) are also a result of fluoride toxicosis (Choubisa [2010a,](#page-9-0) [b,](#page-9-0) [2012b](#page-9-0)).

In a recent study, a new form of industrial dental fluorosis was also observed and reported in domesticated goats grazing in the vicinity of superphosphate factories located near Udaipur City of Rajasthan (Choubisa [2015](#page-9-0)). Interestingly, this unique form of dental fluorosis is characterized with the presence of a single large deep-brownish spot surrounded by alternate light and deep stained thin layers and located towards the upper (incisal 3rd) region of each incisor (Fig. 14). A similar pattern of industrial dental fluorosis has also been detected and reported in cattle grazing in the vicinity of aluminum and fertilizer units in Kerala (Kumar and Aravindaksha [2015\)](#page-10-0). Appearance of such type of dental fluorosis in domestic animals is definitely due to an irregular exposure to a variable concentration of fluoride which is possible in the case of industrial fluoride emission (Choubisa [2015](#page-9-0)). The extent to which deposited fluoride persists on the herbage or feed (leaves, pods, flowers, and small fruits) of the animals may vary between the rainy and dry seasons. In the rainy season, most of deposited fluoride may be washed off the contaminated herbage so that a low level of fluoride exposure is possible. In contrast, during the dry season, cleaning of the vegetation by rain will be less frequent and the concentration of fluoride will increase steadily due to the regular accumulation of fluoride emitted from the fertilizer plants. Another possible reason for the discontinuous fluoride exposure is the seasonal replacement of leaves, fruits, pods and flowers by new ones, which will initially have a low fluoride content. However, the appearance of such unique pattern of dental fluorosis distinguishes it from hydrofluorosis and can probably be considered as a bio-marker for chronic industrial fluoride poisoning or pollution/contamination.

Whatsoever, dental disfigurement or fluorosis is also important because it reduces the life-span of animals. When these dental lesions become severe enough to cause difficulty in grazing and mastication, the animals die at a young age from hunger and cachexia (WHO [1970](#page-10-0); Wang et al. [2003\)](#page-10-0). Nevertheless, the death of animals at an early age has economic consequences for herdsmen.



Fig. 14 Industrial dental fluorosis in mature goat characterized with excessive abrasion of teeth having a deep-brownish spot surrounded by alternate light and deep stained thin layers located towards upper (incisal 3rd) region and appearance of homogenous staining towards lower (gingival 3rd) region of each incisor (Choubisa [2015\)](#page-9-0)

#### Osteal deformities or skeletal fluorosis

It is evident that skeletal fluorosis is more dangerous, very painful and highly significant since it diminishes the mobility of fluorotic man and animals in very early age by producing varying changes in the bones such as periosteal exostosis, osteosclerosis, osteoporosis, and osteophytosis (Choubisa [1996,](#page-9-0) [2012b,](#page-9-0) [d](#page-9-0); Choubisa and Verma [1996](#page-9-0)). These progressive changes are visible in the radiographs or X-rays of human cervical spines (Fig. 15), bones of thorax region (Fig. [16\)](#page-6-0), limbs (Figs. [17](#page-6-0)–[22](#page-6-0)), and pelvic region (Fig. [23](#page-7-0)). The excess accumulation of fluoride in muscles also diminishes or restricts the bone movement which leads to crippling (in humans) or lameness (in domestic animals). Neurological complications such as paraplegia, quadriplegia (Fig. [24\)](#page-7-0), and genu-valgum or genu-varum (Fig. [25](#page-7-0)) syndromes in human



Fig. 15 Radiograph of cervical spines of subject having severe skeletal fluorosis showing progressive osteosclerosis, osteophytosis, and calcification of thyroid cartilage (Choubisa [2012c](#page-9-0))

<span id="page-6-0"></span>

Fig. 16 Radiograph of chest of subject having severe skeletal fluorosis showing a peculiar contrast between the chalky-white bony cage and translucent lungs, irregular thickening of ribs and clavicles and calcification of muscles (Choubisa [2012c\)](#page-9-0)

beings are the resultant of severe skeletal fluorosis. The X-ray of radius and ulna bones is the most ideal in the diagnosis or confirmation of skeletal fluorosis due to the early appearance of the calcification in the interosseous membrane between these two bones (WHO [1970\)](#page-10-0). To the best of our knowledge,

in India, except experimental study on goats (Kant et al. [2009\)](#page-10-0), no other radiological studies were conducted so far on natural skeletal fluorosis in domestic animals. Although intermittent lameness, swollen joints, debility, mortality, wasting of body muscles, and bony exostosis or lesions in the mandibles, ribs, metacarpus, and metatarsus regions are well recognized in the fluorosed animals (Fig. [26](#page-8-0)) (Choubisa [2010a,](#page-9-0) [b,](#page-9-0) [2013a\)](#page-9-0).

In India, a variable prevalence of industrial dental and skeletal fluorosis in man and animals has been reported from Chhattisgarh (Sharma and Pervez [2004](#page-10-0)), Gujarat (Desai et al. [1988](#page-10-0)), Kerala (Kumar and Aravindaksha [2015\)](#page-10-0), Orissa (Rao and Pal [1978](#page-10-0); Naik and Samal [1985,](#page-10-0) [1986](#page-10-0); Samal and Naik [1988,](#page-10-0) [1992](#page-10-0); Saho and Ray [2004](#page-10-0); Mishra and Pradhan [2007\)](#page-10-0), Rajasthan (Patra et al. [2000](#page-10-0); Choubisa [2015;](#page-9-0) Choubisa and Choubisa [2015\)](#page-9-0), Uttar Pradesh (Swarup et al. [1998\)](#page-10-0), and western India or north of Bombay (Bhawsar et al. [1985a](#page-9-0), [b\)](#page-9-0). The highest prevalence, 55.2 % of industrial dental fluorosis in children living in the vicinity of the aluminum factory in Orissa (India) was reported by Samal and Naik ([1988](#page-10-0)). Recently, in another study conducted in western Orissa, 14 to 22 % incidence of dental fluorosis was also reported (Mishra and Pradhan [2007](#page-10-0)). In a large scale survey having sample size of 7059 individuals, conducted in an industrial area of western India, 1202 (17.02 %) individuals revealed the evidence of toxic signs of neighborhood fluorosis (Bhawsar et al. [1985a](#page-9-0), [b](#page-9-0)). In this study, 1655 (23.58 %) individuals of either sex were found to be afflicted with dental fluorosis with varying grades. From the state of Gujarat



Figs. 17–22 Radiographs of the radius-ulna (Figs. 17–19) and tibiafibula (Figs. 20–22) bones of subjects having varying grades of chronic fluoride intoxication showing progressive osteoporosis, roughening and thickening of periosteal surface, periosteal bone formation along the

insertion of tendons, ligaments and the interosseous membranes. Calcification of interosseous membrane of the forearm (Figs. 17–19) as a diagnostic feature of skeletal fluorosis (Choubisa [2012c](#page-9-0))

<span id="page-7-0"></span>

Fig. 23 Radiograph of the pelvic bone with lumbo-dorsal spines or vertebrae of subject having very severe skeletal fluorosis showing diverse progressive changes as sclerosis of the iliac crests and ischial tuberosities, pelvic bones appeared dense with loss of bone detail, osteophytic changes, ossification of sarco-spinous and sarco-tuberous ligaments in advance cases whereas in spines or vertebrae markedly sclerotic with irregular new bone formation, considerable lipping and osteophytosis (Choubisa [2012c](#page-9-0))

(India), 35.3 % tribals residing nearby the fluorspar mines were also found to be afflicted with mild to severe dental fluorosis (Desai et al. [1988\)](#page-10-0). From Rajasthan, 8.33 % goat animals and 55.5 % villagers exhibited prevalence of industrial dental fluorosis (Choubisa [2015](#page-9-0); Choubisa and Choubisa [2015\)](#page-9-0). Extensive epidemiological studies on industrial skeletal fluorosis in different states are required to know the status of impact of industrial fluoride pollution which also helps in making of national health policy for man and animals.

The variation and severity of fluorosis or chronic fluoride intoxication in man and animals living in any geographical provinces can be attributed to a number of factors such as the duration of fluoride exposure and its concentration and intake frequency, age, sex, chemical constituents in drinking water, habits, nutrition or food constituents, environmental factors, besides the individual susceptibility and biological response or tolerance, and genetics of an individual (Choubisa et al. [2007](#page-9-0), [2009,](#page-10-0) [2010](#page-10-0), [2011b,](#page-10-0) [c](#page-10-0); Choubisa [2010c,](#page-9-0) [2012c,](#page-9-0) [2013b](#page-9-0), [c\)](#page-9-0). But authors believe that variation in the prevalence and severity of industrial and/or neighborhood fluorosis is greatly dependent on the distance between the potential source of fluoride emission and the subject or animals. The distance from fluoride emission source definitely reduces the frequency or chances of fluoride exposure, which in turn decreases the prevalence and severity of fluorosis. Such observations were also taken by Samal and Naik ([1992\)](#page-10-0), Patra et al. [\(2000\)](#page-10-0), Saho and Ray ([2004](#page-10-0)) and Desai et al. ([1988\)](#page-10-0) in their studies conducted in industrial



Figs. 24–25 Cases of severe skeletal fluorosis showing common deformities: kyphosis, invalidism, para and quadriplegia, genu-varum syndrome and crippling (Fig. 24) and crossing or scissor-shaped legs (Fig. 25). (Choubisa [2012b\)](#page-9-0)

<span id="page-8-0"></span>![](_page_8_Picture_1.jpeg)

Fig. 26 Emaciated <2-year-old calf afflicted with severe skeletal fluorosis. Note wasting of body muscles and bulging lesions on legs (Choubisa et al. [2012\)](#page-10-0)

areas. Height of chimney and weather condition, such as the persistence of the prevailing wind (pattern and direction) and amount of rainfall are also important contributory determinants for the genesis and severity of neighborhood and industrial fluorosis in man and animals, respectively (Choubisa [2015;](#page-9-0) Choubisa and Choubisa [2015](#page-9-0)).

# Diverse adverse changes in soft tissues or non-skeletal fluorosis

In fluoride endemic provinces the most common health complaints such as gastrointestinal discomforts (nausea/loss of appetite, gas formation or bloating, pain in the stomach/colic, constipation, intermittent diarrhea, headache, etc.), frequent tendency to urinate (polyuria)/itching in the region, excessive thirst (polydipsia), extreme weakness/muscles weakness, allergic reactions, asthma, bronchitis with violent cough, nasal irritation, irregular reproductive cycles, abortion, still birth etc., are the resultant of chronic fluoride intoxication (Susheela et al. [1992,](#page-10-0) [1994](#page-10-0); Choubisa [2010a](#page-9-0), [b,](#page-9-0) [2012a,](#page-9-0) [2013a,](#page-9-0) [b\)](#page-9-0). In a recent study conducted on the 462 employees of aluminum industry located in the northeastern part of the state of Uttar Pradesh, many of them revealed non-skeletal manifestations which indicate that employees are suffering with industrial fluoride poisoning (Susheela et al. [2013\)](#page-10-0). The above mentioned health complaints mostly disappear within short period of 7–10 days, after cessation of fluoride exposure. Significance of these complaints is that these can indicate the

earliest sign of fluoride poisoning (Susheela et al. [2013\)](#page-10-0). But these health problems may be possible because of other reasons. However, for the confirmation of fluoride poisoning, besides the estimation of fluoride content in urine and blood serum, the appearance dental and skeletal changes are more important and highly significant for the confirmation of industrial fluoride intoxication.

# Bio-indicators and bio-markers for chronic fluoride intoxication

Bio-indicators for chronic fluoride poisoning should have less resistance or tolerance but should have a greater susceptibility to fluoride exposure and give early signs of fluoride poisoning. Recently, a study was conducted in different animal species of 2,747 mature and 887 immature domestic animals residing in areas with naturally fluoridated (>1.5 ppm) drinking water. These animals included buffaloes, cattle, camels, donkeys, horses, goats, and sheep. Among these animals, immature ones were found more susceptible to fluoride toxicity (dental fluorosis). However, bovine calves are found to be ideal bio-indicators as these showed an early sign of fluoride poisoning in the form dental fluorosis (Choubisa [2014\)](#page-9-0).

A bio-marker or biological marker generally refers to a measured characteristic which may be used as an indicator of some biological state or condition. The term occasionally also refers to a substance whose presence indicates the existence of living organisms. These bio-markers are often measured and evaluated to examine normal biological processes, pathogenic processes or pharmacologic responses to a therapeutic intervention. Fluoride content in the environmental samples such as forage and fodder indicates the persistent of fluoride pollution in the environment. However, fluoride contents in biological (milk, urine, blood serum, nails, teeth etc.) samples are also better bio-markers for fluoride poisoning in man and animals in contrast to morbidity and mortality. Nevertheless, the presence of fluoride in urine is the most ideal method for the indication of chronic fluoride intoxication (Boddie [1947;](#page-9-0) Samal and Naik [1992](#page-10-0); Swarup et al. [2001\)](#page-10-0). Authors believed that more comparative studies are still needed for the consideration of ideal bio-indicators/bio-markers for chronic fluoride poisoning or pollution/contamination.

# Conclusion

In India, waterborne hydrofluorosis in both man and domestic animals has been widely studied almost in every state. In contrast, reports on industrial fluoride pollution or contamination along with industrial and neighborhood fluorosis in residents and animals living in the vicinity of fluoride discharging industries are very limited. To ascertain the status of impact of <span id="page-9-0"></span>chronic industrial fluoride exposure, more epidemiological studies on industrial fluoride intoxication are needed. Such studies are also helpful in the making of health policy at both state and national level. It is true that most of the industries are located in the remote areas where both forest and wildlife also exist. Therefore, studies pertaining to assess the impact of industrial fluoride pollution on vegetations/plants, wild mammals and birds are also necessary. It is possible that chronic industrial fluoride pollution may cause infertility in wild animals, which in turn decreases their population.

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