On the Toxicity of Fluoridated Water

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Summary
The mechanism by which fluoride's lethal poisoning of man and animals occurs is presented. "Low" level fluoridation of municipal water exhibits well known alterations in teeth and bone structure and calcification of tendons and ligaments. 'Moderate' doses cause spinal deformities and increased hip fracture tendency and kidney and gall stones. Higher levels cause death and are responsible for its major industrial use as a rodenticide. Solubility calculations indicate that fluoride doses required to decrease calcium below physiological blood levels are comparable to those present in poisoned victims' tissues and to those causing decreased beat rates in isolated heart cells in culture. Acute lethal poisoning and many of the chronic 'low' level effects of fluoride are mediated by calcium binding by the fluoride ion.

Introduction
An array of scientific findings indicate that the decision made by many cities as early as 1956 to add fluoride (a rodenticide) to municipal drinking water, as long as the dose is below a certain level (usually 1 part per million, 1 milligram fluoride per liter or 0.05 mM) to decrease the incidence of something as minor as tooth decay, was irrational. We now know that precipitates of calcium fluoride occur in fluoridated water cities when the acidity is low (a pH above 7) depending on the fluoride level used. This causes scaling of water pipes (1) and numerous biological effects in consumers, the extent determined by the acidity and the amount of calcium in the water.

Fluoridated municipal water supplies in the United States have been found to contain fluoride levels ranging anywhere from 0.012 mM to a record lethal accidental 7.5 mM (8). The biologic effects have been diverse, covering the entire above range. In spite of lethal poisonings from municipal water fluoridation programs, the Public Health Service retains its mandate to fluoridate all U.S. cities as soon as possible and to reach out to other cities throughout the world in an effort to minimize tooth decay while fluoridating the blood of the water consumer as though this were an acceptable alternative to topical fluoride or to addition of fluoride to one's own consumed water.

Unfortunately, in 1992 at the mouth of the Yukon River in Hooper Bay, Alaska the unthinkable

occurred. In what is considered an accident, an entire village was poisoned by its own fluoridated water supply when the system malfunctioned. This represents the first 'experiment' in which human beings were exposed to lethal doses of fluoride. Blood samples were measured for incorporated fluoride and calcium ion and provided much pathologic information on the effects of high doses of fluoride assimilated from municipal drinking water supplies (8). 296 residents were severely poisoned with one fatality. Most had heart malfunction-associated symptoms and severe gastrointestinal pain.

It is now understood that the conversion of fluoride ion into HF, hydrofluoric acid, occurred in the stomach due to the stomach acid HCl at pH 3 and the HF caused the intense pain. HF cannot be stored in glass since it dissolves the container; it also dissolves leather and skin. Also blood calcium levels dropped to 1/3 of normal in one victim, causing a heart attack and the loss of his life. Although the authors of the study were uncertain whether the fluoride itself caused the effect directly or rather was due to its known ability to precipitate magnesium or calcium ion, our recent computations indicate that low blood calcium is responsible for the lethal effect of acute fluoride poisoning, as indicated below.

Precipitation of calcium fluoride into peoples' bones, tendons and ligaments (9) occurs depending at typical doses added to municipal water. The condition known medically as fluorosis is associated as expected with spinal rigidity and bone fragility (2), the severity depending on the fluoride level present in the blood and for how long.

If fluoride exposure is sufficiently high or prolonged, formation of kidney and gall stones is known to occur, due to the low solubility of calcium fluoride (0.04 mM at pH 7 at room temperature) (4,6). People with hyperparathyroidism or osteosclerosis are more susceptible in this regard to chronic consumption than others since the calcium fluoride deposits in the soft tissues more efficiently because of lack of sufficient binding sites in bones for it (1).

Interestingly, in children raised on fluoridated water, teeth themselves are more rigid while at the same time may be somewhat more resistant to cavities, but no such effect on adult teeth occurs according to many sources (1, chap. 39, p. 896). Thus fluoridation of adult blood is unnecessary and indeed useless for this purpose.

The dean of Tulane University in New Orleans indicated that fluoridated water consumption at certain doses eventually causes gum disease and for this reason New Orleans water was not fluoridated at the time Chicago and New York and other cities approved it (1). Also, in 1960 under oath in Chicago, the researcher for the Public Health Service who started the fluoridation idea admitted that his data constituting the scientific basis for fluoridation were invalid, shattering its foundation (1). The original observation that people consuming water in Texas that happened to have fluoride into it also had whiter teeth than usual was insufficient to justify mass fluoride addition to other public water supplies, since no one was cognizant of the coexistence of other unhealthful effects that also occurred.

The effects of fluoride are subtle enough to go unnoticed for most people at the levels of fluoridation used currently in Southern California (0.012 mM)(Vallecitos Municipal Water district handouts) and at the increased levels proposed to be used. But since fluoride is converted completely in the stomach to hydrofluoric acid (5), the most corrosive substance known to man, it is likely that consumption of fluoride at levels used in some cities is associated with ulceration of gastric and duodenal tissue (where the pH has yet to return to basic values that occur in the middle intestine). And many report evidence in rats that it eventually causes cancer (1),.

Some argue these effects are unimportant if the municipal water supply maintains very low levels of fluoridation; but the longer the consumption occurs for an individual and the more elderly the person with less cell division occurring in the gastric mucosa, the more overt symptoms become. Individuals with ulcers or heartburn are not good candidates for the long term consumption of water containing fluoride, particularly at doses allowed by the Public Health Service (2-4 mg/L, 0.1-0.2 mM)(VWD handouts). These high doses can be dangerous depending on the amount of water consumed, the individual's own body chemistry, and the ionic composition and pH of the particular cities' water that would be fluoridated to this level.

We here determine whether and to what extent blood levels of calcium may be affected by various fluoride doses that are known to occur in the blood of fluoridated water consumers to attempt to determine its modes of action. Our calculations are consistent with the notion that fluoride's lethal

effects on the heart are due to low blood calcium subsequent to saturation of body fluids with fluoride at its known low solubility in the presence of physiologic levels of calcium.

Analytical Results and Discussion

Sublethal poisoning occurs at 0.1-0.2 mM fluoride in blood (3,7) and lethal poisoning occurs in the 0.2 to 0.6 mM range due to heart failure (3). We investigate the possibility that the margin of safety is so slight between unnoticed effects (0.02-0.05 mM) to sublethal (0.1-0.2 mM) and lethal poisoning (0.2-0.6 mM) is because below the critical concentration of fluoride in the blood that causes precipitation of calcium fluoride only chronic, often unnoticed effects would occur. Much like being near a hot electrical wire, one can coexist next to it for lifetimes without any difficulties. But one false movement too close to the wire would be a disaster.

With this in mind, we calculated the concentration of fluoride that would cause calcium fluoride precipitates to first form from the known solubility product constant (Ksp) for calcium fluoride (Ksp = 3.4 x 10-11 (6)) and the known concentration of calcium ion in normal human blood (3 mM) (5). The computed dose is 0.1 mM. Here the concentration of fluoride is: $[F^-] = (Ksp/[Ca^{2+}])^{1/2}$ from the definition of the solubility product constant for insoluble salts where CaF_2 $Ca^{2+} + 2\ F^-$ and $Ksp = [Ca^{2+}][F^-]^2$ (see Table I). The concentration of fluoride where the blood calcium level would be lowered to the lethal low level of about 1 mM is 0.2 mM fluoride.

In Table I the calculated calcium levels that would coexist in fluid with a given fluoride level from solubility considerations are compared with actual measurements of blood levels of calcium and fluoride ion in the lethal poisoned human victim from Hooper Bay, Alaska. Note the good agreement between theoretically calculated fluoride levels, that should lower blood calcium ion to levels below normal, with the actual calcium and fluoride ion levels measured in the blood of this human victim poisoned with fluoridated municipal water in Hooper Bay.

Also note the below-normal calculated calcium ion level that would coexist with fluoride doses found to slow heart cell beat rates in detailed in vitro experiments (10). Isolated beating heart cell preparations from mammals exhibit beat rates that are proportional to the calcium ion level in the incubation medium from .3 - 3 mM. Calcium chelating agents EGTA and EDTA and the calcium binding site competitor La^{3+} ion completely block excitation-contraction coupling in intact beating hearts and in isolated cell preparations (11). Further, addition of fluoride to beating heart cell preparations slows beat rates in a dose-dependent manner that Ksp calculations indicate would lower calcium ion levels in the incubation medium (see Table I).

These calculated doses are fully consistent with other published data indicating that tissue levels of fluoride in poisoned people are in the 0.2 - 0.4 mM range (5). Also the known human lethal dose is 1-5 grams per adult taken at one time acutely (3,5). Since the average adult contains about 43 liters of body fluid this corresponds to a concentration of fluoride of 0.5 mM in such a case of instant acute poisoning.

Wang, Zhang and Wang also found the heart cell beat rate in cultured cells in well-controlled experiments progressively slows with increasing fluoride levels in a regular, concentration-dependent manner (10). Unlike skeletal muscle, cardiac muscle requires extracellular calcium ion from the bloodstream to couple electrical excitation of the cell membrane with contraction of cardiac muscle fibers (11). Each time the heart contracts, calcium fluxes into the heart cells from the extracellular fluid (at 3 mM calcium ion normally). When the heart relaxes, the calcium is pumped back out of the cell, allowing the fibrils to relax. Lowered extracellular calcium ion levels block contraction of the heart.

These data together suggest that the mechanism by which fluoride ingestion is lethal is by causing hypocalcemia and blockage of heart contractions. Fluoride levels in blood below 0.1 mM do not lower calcium ion below normal as no precipitate yet forms in the blood at this or lower doses. But the instant fluoride exceeds this amount to any degree, calcium ion precipitates and the blood level is lowered, unable to support normal heart function.

Fluoride acts as an enzyme inhibitor for all enzymes requiring calcium for function by binding the ion and is used routinely to block sugar metabolism in red blood cells for clinical laboratory analyses of blood specimens. Fluoride also attaches to calcium anywhere this ion is concentrated throughout the body, including teeth, bones, ligaments, skeletal muscle and brain. But the most crucial function

requiring calcium that is fluoride-sensitive is the mechanism of contraction in normal beating hearts.

That extracellular calcium is an obligatory requirement for heart cells to undergo contraction after electrical excitation is well known. Heart cells do not have well-developed sarcoplasmic reticulum to store calcium for this purpose as does all skeletal muscle, which does not exhibit this extreme sensitivity to changes in blood calcium level. The cellular uptake of calcium occurs during the plateau phase of the cardiac action potential and extracellular calcium is necessary for the development of contractile force (11). The strength of contraction (inotropic state) of the heart depends on calcium, where half maximal contractility occurs at 0.5 mM calcium outside cells (12).

It is also possible that chronic 'low' level biologic effects of fluoride are also mediated exclusively by binding and sequestration of calcium. Prior to levels of calcium in the blood being lowered (below 0.1 mm fluoride), regions in the body enriched in calcium would still precipitate calcium fluoride, as in bone, teeth, ligaments and brain. The usual physiologic response to such an insult is to increase levels of hormones such as calcitonin to mobilize calcium from bone to fight the sequestration. At higher fluoride doses, precipitates may be directly responsible for the known formation of gall and kidney stones in fluoridated consumers.

The current level of fluoride in Southern California drinking water is 0.25 mg per liter or 0.012 mM. The blood level is typically in consumers about 1/5 to 1/8 the water level. This is below the solubility for calcium fluoride at normal body pH, temperature and prevailing body fluid calcium levels, and it is easy for many to assume the information in this manuscript is irrelevant. But some cities use up to 1 or 1/5 mg/L (0.05-0.075 mM) or the Federal allowed ceiling of 2-4 mg/L (0.1-0.2 mM) and are near or at the maximum level that would just begin precipitation of calcium, with hypocalcemia, unless the city water happened to have so much calcium in it that it precipitated as the fluoride preventing the fluoride added from entering one's blood at that level. The finding that fluoridated cities generally have increased per capita heart attack rates is consistent with this discussion.

The fluoride level that would precipitate calcium from Southern California water (where calcium ion is about 2 mM) would be 0.14 mM fluoride. So before we could reach fluoride levels approaching the Federal ceiling in water it would precipitate calcium from our drinking water first. To maintain a higher level of fluoride than 0.14 mM would be expensive, requiring addition of enough to precipitate the calcium in the water first. More would be required on top of that amount to increase fluoride to a higher desired level. Fortunately this would be very difficult.

Adding sodium fluoride to public water is paid for by taxpayer adults who will not reap any measurable benefits from it. It takes resources, time, chemicals and machinery to continue to add it to drinking water. It is putting the water district in charge of drugging the public and for something as innocuous as a cavity rather than for serious effects such as infectious illness for which we have properly chosen chlorination, with the much less electronegative halogen.

It is not in keeping with a free society or with proper health care practice to impose these risks associated with fluoridating the blood of people, livestock, and pets, and also all agricultural products, not to mention our lawns and gardens, compared to the less significant problem of perhaps having tooth decay. Tooth decay should be minimized more efficiently and safely if desired with addition of fluoride products to children's teeth carefully without swallowing or better yet by simply brushing more vigorously and regularly. After the death of the Brooklyn, New York boy in the dentist chair when fluoride gel was swallowed, and after the Hooper Bay, Alaska incident, it is clear that our blood is more important than concern for cavities. Teeth are replaceable but lives are not. In keeping with the Hippocratic oath, no physician reserves the right to medicate anyone without their permission, and all patients must remain free to withdraw from drug or other treatment programs at any time. Forced fluoridation in public water supplies ironically constitutes a reversal of these Public Health Service policies. The easy way - fluoridate through the bloodstream by drinking - is unnecessary (since topical application is possible) and criminal (in light of the above findings). Proper dental hygiene is much safer and achieves the desired result anyway. The notion recently publicized that 'antifluoridationists' are similar to earlier critics of smallpox vaccination is inconsistent with the facts that 1) smallpox is lethal and could not be prevented without blood vaccination, but 2) cavities are not lethal and can be prevented with proper hygiene and if necessary the bacteria that cause caries in the first place can be quickly destroyed with simple methods such as hydrogen peroxide washings, etc. without loss of life.

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Table I

Effects of F- on Blood Ca2+ Concentrations*

Blood [Ca2+] Blood [F-] (mM)

3.0 0.10 (F- , calculated from Ksp, for 1st precipitation of normal blood Ca2+)

1.3 0.48 (human blood measurements, Hooper Bay, Alaska from lethal dose victim)

1.1 0.15 (Ca2+ calculated from Ksp for F- dose lowering heart cell beat rate 17%)

1.0 0.20 (F- calculated from Ksp to lower blood Ca2+ to 1 mM)

0.4 0.30 (Ca2+ calculated from Ksp for F- dose lowering heart cell beat rate 27%)

*Some cities recommend 01. - 0.2 mM fluoride be added to drinking water. Typically 1/5 or so of the water fluoride level is the consumers' blood fluoride level (as long as there are no accidents, equipment malfunction as in Hooper Bay disaster, or miscalculated doses added).

As for any insoluble precipitate, the Ksp solubility product constant determines the concentration in solution of the ions that dissolve from the salt. For calcium fluoride where CaF2 Ca2+ + 2F-, Ksp = [Ca2+][F-]2 = 3.4 x 10-11. This relation was used to calculate F- levels for a given Ca2+ level or Ca2+ levels for a known F- level. Other measurements in the table were from actual blood samples drawn from Hooper Bay, Alaska victims where fluoridated municipal water for which machinery malfunctioned poisoned 296 residents. Not mentioned is the increased thirst associated with heavily fluoridated water, a biologic response to this insult that was up to that time unknown.

The solubility of calcium fluoride changes somewhat with temperature and pH. It is slightly more soluble at body temperature (37oC) than room temperature but since it also decreases in solubility with increasing basicity, we here estimate the solubility in blood at about the published value at pH 7 for water at room temperature because the slight increase it actually has is offset by the higher pH of blood, at 7.4.