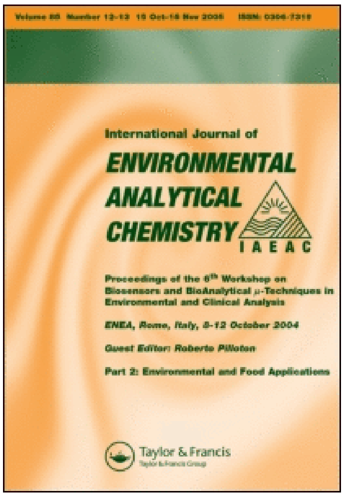

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Determination of Fluorine in Tea

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A simple method is presented for the determination of the fluorine content of tea by using a 0.5–1.0-g sample. Fluorine is fixed by means of calcium oxide during the destruction of organic matter and then converted to soluble fluoride by fusion with sodium hydroxide. The fluoride is determined titrimetrically after steam distillation of fluosilic acid. Analysis of 16 varieties of tea reveals that fluorine concentration increases with the age of tea leaves upon harvest and that commercial teas contain fluorine in the range between 40 and 120 ppm. Large amounts of fluorine-containing compounds in the environment lead to extremely high fluorine values in the leaves of *Camellia sinensis*, suggesting that it may be used to monitor the extent of pollution.

INTRODUCTION

Fluorine has long been recognized as an environmental problem. In the early 1930s it was found that natural water with high fluorine contents caused black teeth, while drinking water with a fluoride level of 1 ppm is known to prevent dental caries.[1,2] The fluorine intake also can be derived from food.[3,4] Since tea is the standard beverage for nearly one-half of the world's people, it was deemed worthwhile to investigate the fluorine content obtained from various sources. The literature on the composition of tea has been concerned with organic constituents[5–9] and trace metals;[10] there is no report on fluorine in tea. In the present paper, a simple method to determine fluorine in 0.5 to 1.0 g of tea is described, and the analytical results obtained from 16 varieties of tea are interpreted.

EXPERIMENTAL

Equipment and Reagents

The apparatus and reagents for H_2SiF_6 distillation are given in Ref. 11, and for titration of fluoride in Ref. 12. In addition, the following are required: a nickel crucible, 50-ml capacity; calcium oxide, fluorine-free; sodium hydroxide, fluorine-free; an infrared lamp; a muffle furnace.

PROCEDURE

Accurately weigh 0.5 to 1.0 g of the pulverized tea sample into the nickel crucible. Add 0.1 g of calcium oxide as fixative and char the mixture under the infrared lamp. Then mineralize in the muffle furnace at 600°C. Fuse the resulting ash in the same crucible with about 5-fold amount of sodium hydroxide, thus releasing completely the bound fluorine in form of soluble fluoride. After cooling, add 10 ml of fluorine-free water and transfer the caustic fusion mixture quantitatively into the distillation apparatus[11] and determine the fluorine content by the method of Ma and Gwirtsman.[12, 13]

RESULTS AND DISCUSSION

The results for fluorine analyses of 16 varieties of tea obtained from different sources are tabulated in Table I. The list includes the two taxa of commercial tea plants, i.e., China tea (*Camellia sinensis*) and Indian tea (*Camellia assamica*). The three types of processed tea leaves are represented, viz., green tea (unfermented), oolung tea (partly fermented), and black tea (fermented). One brand of instant tea was tested also. Duplicate determinations were made. It will be noted that the fluorine content of the commercial teas analyzed falls in the range from 40 to 120 ppm, whereas fluorine is practically absent in the instant tea sold on the market. The accuracy of the distillation and titration procedure is $\pm 4\%$ in the 50-ppm region and $\pm 2\%$ in the 100-ppm region.

Although the number of tea samples in this study is not sufficiently large to draw definite conclusions on the relationship between the fluorine content and the environment of the tea source, some observations may be made as follows.

(1) All varieties of commercial tea leaves investigated contain significant amounts of fluorine. Therefore, besides being a beverage to quench thirst, tea may also furnish a daily supply of fluoride to the tea drinker.

TABLE I

Analysis of fluorine in tea

Tea	Source	F found, ppm (dry weight basis)
Green tea (1)	Mainland, China	67
Green tea (2)	" "	64
Jasmine tea	" "	101
Black tea (1)	" "	118
Green tea (3)	Taiwan, China	42
Green tea (4)	" "	44
Jasmine tea (2)	" "	45
Oolung tea	" "	52
Black tea (2)	" "	73
Lipton tea	American brand	88
Nectar tea	" "	73
Our Own tea	" "	110
Tetley tea	" "	114
White Rose tea	" "	98
White Rose Instant Tea	" "	1.5
Camellia sinensis (old leaves)	Greenhouse, New York	1527

(2) The fluorine content of tea appears to increase with the age of the leaves. Thus, green teas which usually comprise of young foliage show fluorine values lower than black teas which are often made from mature leaves. The high concentration of fluorine in the old leaves of a plant grown in the greenhouse also supports this view.

(3) Although the organic components of tea are known to differ greatly between *C. sinensis* and *C. assamica*,[5, 6] the two species seem to store fluorine in their leaves at about equal levels. All tea samples from China belonged to *C. sinensis*, whereas the American brand teas contained mostly *C. assamica*. Since young leaves are rarely used to prepare the American brand teas, they show fluorine values comparable to those of the black teas from China.

(4) The processing of tea leaves by fermentation (oxidation) probably does not change the fluorine concentration. On the other hand, it is interesting to note that the instant tea studied was practically devoid of fluorine.

(5) The low incidence of atherosclerosis among the Chinese people has been ascribed to their tea-drinking habit.[14] It has been reported that an overt tea-drinking population has both lower blood cholesterol[15] and serum lipid values[16] as compared to a coffee-drinking population. Young and co-workers[17, 18] have confirmed that tea decreases concentration of

serum lipids and that this beneficial effect of tea is not due to theophylline or theobromine. It would be of interest to study whether the fluorine-containing substances in tea are related to this factor.

(6) It can be seen from Table I that teas grown in mainland China have higher fluorine contents than those grown on the island of Taiwan. In China, tea is marketed by identifying the locality where it was grown and is not blended. Hence it may be feasible to use fluorine analysis as a means to determine the geographic origin of the tea. This method may be employed to complement the determination of extractives[19] when substitution or adulteration is suspected, and it may be applicable to other plant materials such as spices.[20]

(7) The fluorine found in tea leaves is apparently derived from the soil. When fluoridized water was used in the greenhouse, it produced tea with extremely high fluorine concentration. Based on this observation, tea grown in an environment polluted by hydrogen fluoride or where fluorine-containing compounds are employed as insecticides[21-23] will show comparatively high fluorine content in its leaves. Hence the tea plant may be used to monitor the extent of pollution.

(8) The method described in this paper is applicable to the determination of fluorine in other plant tissues. Since 0.5–1.0 g of the plant material is used in this method, it is considerably more rapid than the procedure described by Horton[24] using a 1–5-g sample or the standard method[25] using a 10–15-g sample.

(9) The visual titration technique employed in this study for the determination of fluoride can be carried out with simple equipment and is convenient for occasional analysis. On the other hand, the spectrophotometric finish is recommended for routine multiple runs. The latter procedure requires the preparation of calibration curves. In a recent investigation of fluorine content of orchard leaves, Schmidt and Paulson[26] used the lanthanum-alizarin complexone spectrophotometric method and found that the calibration curve varied from day to day and also with new batches of reagent.

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