Bull. Natn. Sci. Mus., Tokyo, Ser. D, **12**, December 22, 1986

Fluorine and Phosphate Analysis of Fossil Bones from the Kabuh Formation of Trinil

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Abstract Compact bone samples (n=21) of fossil vertebrate specimens from the lower Kabuh Formation in the Trinil area, East Java, have been analysed for fluorine and phosphate in order to augment comparative data for the fluorine dating of the Trinil hominids which have long been surrounded with controversy in terms of their contemporaneity. Some remarks about the analytical results and their possible bearing on the stratigraphic provenance of Pithecanthropus Skull I and the Trinil femora, are noted.

Introduction

The discovery of the first *Homo erectus* skull, Pithecanthropus Skull I, was made by E. DUBOIS nearly one century ago in 1891 from the left bank exposure of Bengawan Solo in the Trinil area, East Java. One year later an intact femur (Femur I) came to light at a distance of about 15 metres from the skull. Now five specimens are listed as the Trinil femora together with the other four (II-V), which were recognized in 1932 among fossil mammal bones excavated in 1900 from Trinil (the exact locality unknown) and stored in the Leiden Museum (see DAY & MOLLESON, 1973, and OAKLEY *et al.*, 1975).

The Trinil femora, in spite of the primitive character of the skull cap, seem to be as evolved and fashioned as they are today; and this has caused repeated doubts about their synchronism. WEIDENREICH (1941), for example, has believed that the Trinil femoral specimens are fundamentally different from the *Sinanthropus* specimens of Choukoutien and that the Trinil femora do not "belong to *Pithecanthropus* but must be attributed to a type closely related to modern man" (Ibid., p. 50). Later BERGMAN & KARSTEN (1952), through fluorine analysis of bones as a relative dating approach, concluded that the Trinil hominid remains are of a similar antiquity, and this support for their contemporaneity has been widely quoted. However, the newly available femoral remains of fossil hominines from East Africa have again given rise to arguments on the association of the Trinil femora with the Trinil calotte (DAY & MOLLESON, 1973; KENNEDY, 1983). In this connection, BARTSTRA (1982, 1983) expresses caution that the DUBOIS collection of fossil vertebrates from Trinil might be a mixture of materials from broad temporal zones, though this possibility is in dispute (SONDAAR, *et al.*, 1983).

Recent reconsideration of the reported chemical data on the Trinil remains has led me to believe that the samples analysed in the study by BERGMAN & KARSTEN (1952) may have suffered from contamination by varying amounts of adventitious matter such as silt, infiltrated minerals and preservative (glue) (see DAY & MOLLESON, 1973), and that the conclusion of BERGMAN & KARSTEN (1952) may also have suffered from lack of adequate comparative data (MATSU'URA, 1984; see Results & Discussion section). MATSU'URA (1984) has also indicated that the extent of fluorination of the Trinil calotte is not uniform with that of the Trinil femora. Yet an essential problem is whether the disparity should be within the fluctuation in fluorination of bones derived from a given stratum or not.

This report gives some data for a discussion of this problem, and is also a note on their implications for the provenance of the human remains from Trinil.

Material

Vertebrate fossils were collected in September 1977 from the Kabuh Formation in the Trinil area during a field survey conducted under the Indonesia-Japan Joint Research Project CTA-41 (see WATANABE & KADAR, 1985). Twenty-one specimens recovered *in situ* were taken for this analytical study from the fossil collection. Of these, 14 specimens derived from gravel bed-1 (KBG1; SOERADI *et al.*, 1985) located at the base of the Kabuh Formation on the left bank of Bengawan Solo in the vicinity of the Pithecanthropus site. This gravel bed corresponds to DUBOIS's layer D (SOERADI *et al.*, 1985) which was reported to have yielded Pithecanthropus Skull I and Femur I (DUBOIS, 1896). The rest 7 specimens derived from gravel facies in the lower Kabuh Formation on the right bank of Bengawan Solo several kilometers upstream (about 1 km westward in a straight line) from the Pithecanthropus site. This gravel facies is supposed to correlate with KBG1 (see above), or a somewhat upper horizon in the lower Kabuh Formation (SHIBASAKI, 1986, personal communication).

The geological and stratigraphical contexts concerned are given in SOERADI *et al.* (1985).

Methods

A small piece of cross section of compact tissue (or dentine) was cut or chipped out of each fossil specimen, and scraped to remove surface dirt. The piece was then well powdered to be homogeneous in an agate mortar.

The pulverized sample was subjected to fluorine and phosphate assays. Fluorine was determined with a fluoride sensitive electrode using a buffer medium to adjust the ionic strength and pH (here, about 5.5) of the sample solution and to overcome metal ion interferences. The buffered system employed has final concentrations of 0.125 M HCl – 0.25 M NaCitrate – 0.033 M NaAcetate – 0.075 M KNO_3 (see MATSU'URA, 1985).

Table 1. Analyses of fluorine and phosphate in compact bone and horn and in dentine of fossil specimens *in situ* from the left bank exposure of the Solo River near the Pithecanthropus site at Trinil.
Horizon: gravel bed (KBG1) lying at the base of the Kabuh Formation.

Specimen	Thickness of analysed part* (mm)	Fluorine %	P_2O_5 %	$100F/P_2O_5$ ratio
elephantid, tusk	>11	1.39	29.6	4.70
vertebrate, long bone	c. 4	1.75	34.9	5.01
vertebrate, tooth	>11	1.80	35.4	5.08
mammal, long bone	1.9	1.85	35.7	5.18
mammal, long bone	1.7	1.93	35.0	5.51
vertebrate, long or short bone	2.6	1.96	35.3	5.55
vertebrate	1.2	2.04	35.8	5.70
vertebrate	3.5	1.92	33.6	5.71
vertebrate	3.8	1.95	32.9	5.93
vertebrate, horn	1.8	2.11	34.6	6.10
cervid, antler	2.8	2.07	33.9	6.11
cervid, antler	2.7	2.10	33.8	6.21
vertebrate, flat bone	1.4	2.21	35.3	6.26
vertebrate	3.5	2.13	31.7	6.72
	Mean	1.99**	34.5**	5.70
	S.D.	0.14**	1.2**	0.57

* Thickness of compact tissue or dentine.

** Excluding data for specimen with 1.39%F and 29.6%P_2O_5 which has suffered a light contamination.

Phosphate was determined by means of normal volumetry (acid-base titration) after precipitation as ammonium phosphomolybdate. For further references on the analytical methods, readers are referred to MATSU'URA (1981, 1982).

Results and Discussion

Listed in Tables 1 & 2 are the resulting data for fluorine and phosphate tests on the Trinil material. Phosphate determination gives a convenient measure of the apatite present in bone sample. Fossilized dentine and compact bones with little infiltration of soil particles or other foreign mineral matter, carry c. 32–36% P_2O_5 content in general (e.g. BROPHY & NASH, 1968; MATSUBARA, 1980; MATSU'URA, 1982 and unpublished data). The Trinil specimens analysed here (Tables 1 & 2) show the same level of phosphate content with an exception which implies a light contamination (elephant tusk, top row of Table 1).

Comparison with Vertebrate Fossils from Sangiran

In the basin of Solo River tributaries about 60 km WSW of Trinil lies an important early hominid site, the Sangiran area, where a sequence of Plio-Pleistocene fossil-

Table 2. Analyses of fluorine and phosphate in compact bone of fossil specimens *in situ* from the right bank exposure of the Solo River in the Trinil area.
Horizon: gravel bed lying at the basal Kabuh Formation or at somewhat upper stratigraphic level.

Specimen	Thickness of analysed part* (mm)	Fluorine %	P_2O_5 %	$100F/P_2O_5$ ratio
vertebrate	>10	1.96	33.7	5.82
vertebrate	>11	2.05	34.6	5.92
vertebrate	6.4	2.19	34.8	6.29
vertebrate	6.8	2.20	33.6	6.55
vertebrate	2.7	2.38	34.2	6.96
tortoise, shell	1.3	2.44	33.0	7.39
vertebrate, long or short bone	1.1	2.50	33.1	7.55
	Mean	2.25	33.9	6.64
	S.D.	0.20	0.7	0.68

* Thickness of compact tissue.

bearing formations are exposed (ITIHARA *et al.*, 1985). A large number of vertebrate bones were collected *in situ* from various well-documented stratigraphic horizons during excavations by the CTA-41 project (see WATANABE & KADAR, 1985). To clarify stratigraphic positions of the hominid finds and to develop a chronological framework of the Sangiran hominids, MATSU'URA (1982) conducted a fluorine dating study of key hominid specimens using the systematic collection of animal fossils as a guide series. Fig. 1 represents a series of standard fluorine data for the Sangiran area, with the analytical results obtained for the Trinil vertebrate specimens.

Buried bone takes up fluoride ions from moist surrounding matrices or ground water, and accumulates this element with time at the cost of hydroxyl ions in the apatite structure. Thus the fluorine analysis of bone provides relative dating information. In the case of fossil remains from the Sangiran area, their fluorine content serves as an index to identify their original stratigraphic provenance since it has revealed a significant difference between fossils from different layers (MATSU'URA, 1982). However, the average content of fluorine fails to vary monotonically with age (Fig. 1) but instead reflects the variable hydrological and geochemical influences of burial media (MATSU'URA, 1982). As will be seen from Fig. 1, the fluorine content of compact bones from the gravel bed (KBG1) at the basal Kabuh of Trinil compares well with that of compact bones from the Grenzbank zone in Sangiran. This finding is in harmony with the observation that KBG1, 'a conglomerate with components of andesite and limestone fragments, has a similar appearance to the fossil-bearing bed, "Grenzbank" of the basal part of the Kabuh Formation in the Sangiran area' (SOERADI *et al.*, 1985, p. 53).

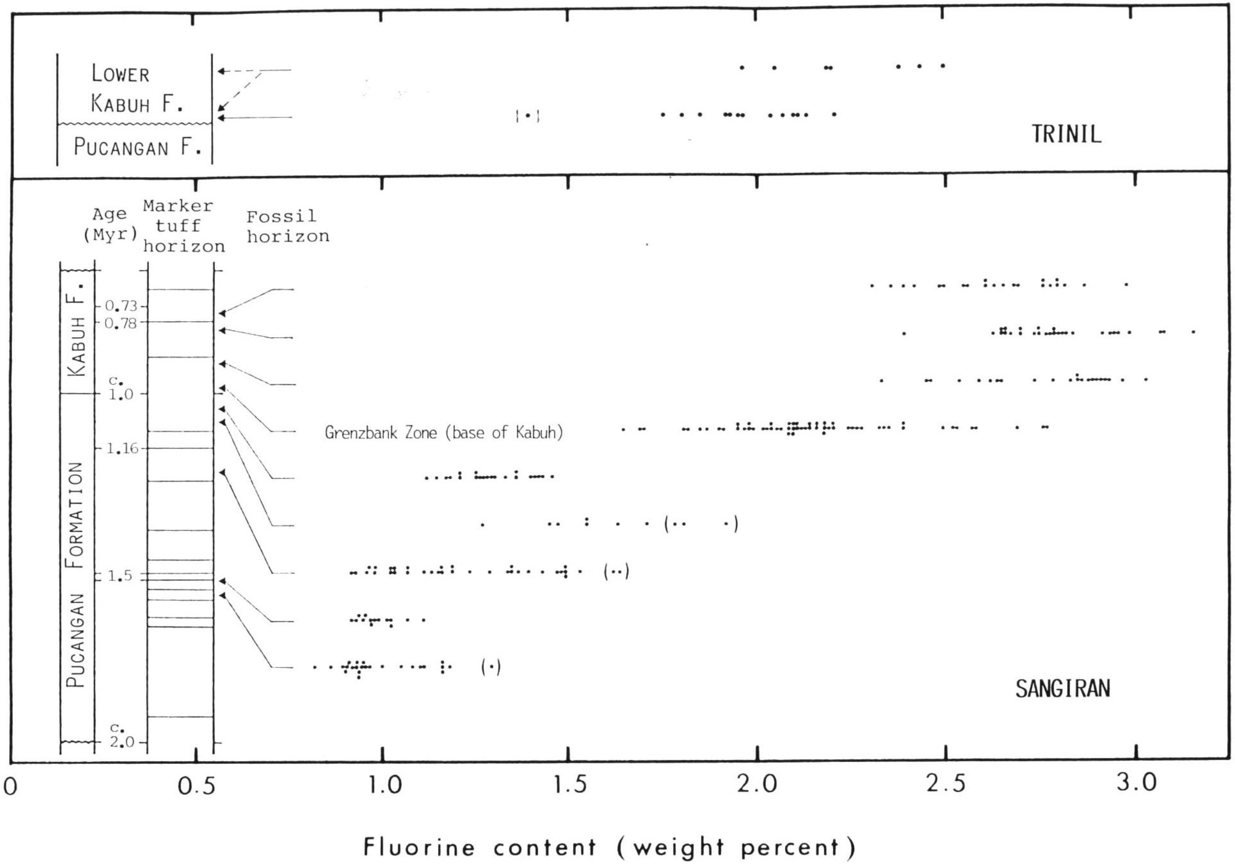

Fig. 1. Comparison of fluorine content in *substantia compacta* of bone and horn, and in dentine from fossil vertebrate remains of the Trinil area (this study) and the Sangiran area (Matsu'ura, 1982) in Java. Data in parentheses are those for samples less than 1 mm thick in cross-section of compact tissue; and one in braces for a lightly contaminated tusk sample. The stratigraphical contexts and absolute ages are based on the work by the CTA-41 team (e.g. Watanabe & Kadar, 1985).

Table 3. Fluorine and phosphate contents of fossil hominid remains from Trinil.

Code No.*	Designation	Fluorine % (1)	Fluorine % (2)	P_2O_5** % (2)	$100F/P_2O_5$ ratio (2)
T2	P. Skull I	1.22, 1.20	1.14	27.8	4.1
T3	Femur I	1.05, 1.12	1.14	19.7	5.8
T6	Femur II	1.01, 1.01	0.72	11.6	6.2
T7	Femur III	1.34, 1.43, 1.35	1.84	32.3	5.7
T8	Femur IV	1.38, 1.43	1.8	31.0	5.8
T9	Femur V	1.10, 1.02	1.79	30.9	5.8

(1) From BERGMAN & KARSTEN (1952). (2) From DAY & MOLLESON (1973).
* British Museum's Catalogue No.; T=Trinil.
** Calculated from F content and $100F/P_2O_5$ ratio.

Comment with reference to the Trinil hominids

After the analytical study by BERGMAN & KARSTEN (1952), DAY & MOLLESON (1973) report the results of repeating fluorine test on the Trinil femora and calotte, which are accompanied by phosphate analyses (Table 3). The accompanying phosphate measurements give evidence that the tested samples of the Trinil calotte and Femora I & II were contaminated to a great or small extent by extraneous substances. MATSU'URA (1982) has checked that a correction procedure on the basis of phosphate determinations permits comparison of the fluorine content between cancellous or contaminated regions and dense cortical regions of bones. Taking the fluorine/phosphate ratio ($100\times \%F/\%P_2O_5$) is one presentation for eliminating the contamination problem (e.g. OAKLEY, 1951, 1980), and used hereunder.

The five Trinil femora have exhibited uniform fluorine/phosphate ratios (Table 3), clustering from 5.7 to 6.2 (when $P_2O_5=34\%$, $F=1.9$–2.1), while the calotte is clearly out of this range with fluorine/phosphate ratio of 4.1 (when $P_2O_5=34\%$, $F=1.4\%$); although MOLLESON (1981) has regarded the fluorine content of the calotte and femora as similar. According to BERGMAN & KARSTEN (1952) the fluorine content of the femora ranged from 1.01 to 1.43, and that of the calotte was included in this range (Table 3). This outcome may have arisen from the probable contamination of varying degrees to the samples involved.

As mentioned above, the fluorine/phosphate ratios of the Trinil femora focus around 6.0. These values fall well within the range of fossil bones (Table 1, Fig. 2) from the basal Kabuh bed that corresponds to DUBOIS's layer D (see Material section). Further assays for other elements of geochronological or geochemical significance on the Trinil femora along with an adequate 'pilot series' (see OAKLEY, 1951) of Trinil materials from known horizons, would be necessary for examining the possibility, if any, of the femora being intrusive from (an) upper horizon(s).

On the other hand, the fluorine/phosphate data on the Trinil calotte (4.1) does not locate in the coverage of animal fossils from Trinil (4.70 to 7.55: Tables 1 & 2). Previous works have presented three data on the fluorine/phosphate ratio for Trinil bones,

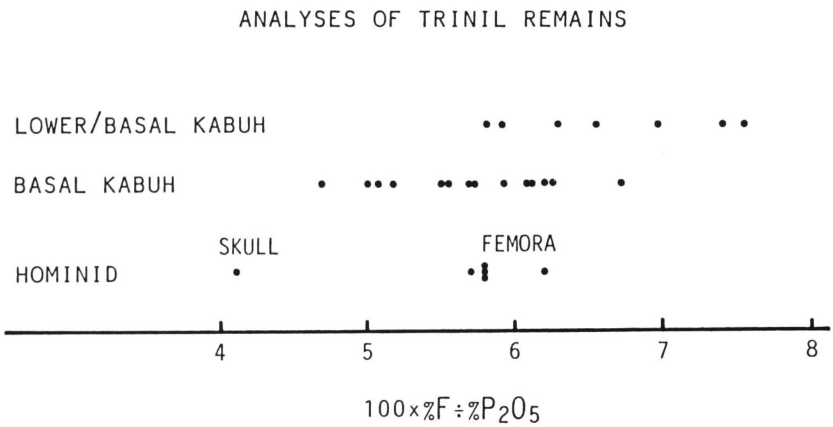

Fig. 2. Ratio of fluorine to phosphate for animal and human fossils from the Trinil area. Exact figures are given in Tables 1–3.

that is, 4.8 for a *Stegodon* tibia (BEMMELEN, 1897), 6.9 and 6.2 for a *Bibos* mandible and an *Axis* antler respectively (DAY & MOLLESON, 1973); they fall within the range of data obtained in this study (Tables 1 & 2). Again the human calotte is outlying (Fig. 2). The lower level of fluorination could suggest the possibility of the calotte deriving from an uppermost/upper part of the Pucangan Formation on comparison with the data on Sangiran materials for a tentative reference (see Fig. 1). The fluorine content (or its ratio to phosphate), however, does vary though on a minor scale within a given specimen. If we take account of such a possible fluctuation (± 2 S.D.) of the fluorine/phosphate ratio in a specimen as expected from MATSU'URA (1982), the calotte could locate just on a lower limit of the fluorine/phosphate ratios for bones from the basal Kabuh of Trinil (Table 1). It is hoped to make a reassay on Pithecanthropus Skull I using a less-contaminated sample taken from compact regions of the specimen.

DAY (1984) reports the use of a new technique termed Energy Dispersive Microanalysis for detecting various elements, and preliminary results on samples taken by K. P. OAKLEY from the Trinil hominid remains. However, some minor elements such as Si, Fe and Mn can occur in the form of separate minerals filling voids and crevices in the bone (e.g. PARKER & TOOTS, 1970), and should have difficulties in assessing the significance of the analytical data.

The clarification of the provenance of the Trinil hominids demands further field study in that area, especially to collect fossil specimens from various well-documented horizons, and laboratory work.

Acknowledgements

I wish to express my deep gratitude to Professor Emeritus Naotune WATANABE for his valuable advice and encouragement, to Dr. Tatsuo SHIBASAKI for kindly responding to my questions concerning the provenances of the fossil vertebrate specimens used in this study, to Dr. Mitsuru AIMI for his helpful suggestions. Grateful thanks also go to Dr. Hajime SAKURA and Dr. Bin YAMAGUCHI for their interest and reading the manuscript, to Miss Akiko NAKATSUKA for her help in preparing Figures 1 & 2.

This work is a continuation of the study made as a part of the Indonesia–Japan Joint Research Project CTA-41, co-headed by Dr. Naotune WATANABE and Dr. Darwin KADAR. The expenses of this work were in part defrayed by the Grants in Aid for Fundamental Scientific Research from the Ministry of Education, Science and Culture, Japan (No. 59740426).

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