

A Chronological Framing for the Sangiran Hominids

—Fundamental Study by the Fluorine Dating Method—*

By

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Introduction

Palaeoanthropological researches in these twenty years have been revealing the tempo and mode in human evolution. The greater part of recent advance in palaeo-anthropology may be assigned to the Plio-Pleistocene localities in the East African Rift such as Olduvai Gorge, the Koobi Fora region and the Omo basin. These localities are weighty in that within stratified sequences there spanning over a million years series of fossil hominid material have been found through organized explorations and in that the better radiometric ages of the fossil bearing beds have been measured.

The Sangiran area in Central Java, which has yielded many remains of early man amounting to at least 34 individuals since 1936, is the only site in Asia that shows promise to be comparable with the aforementioned hominid sites in Africa. This area, however, presents a special problem that none of the hominid finds have been obtained *in situ* through controlled excavations. These human remains of Sangiran were

* This work constitutes the most part of my doctoral dissertation accepted by the Faculty of Science, The University of Tokyo. Part of this study is reported in MATSU'URA (1983).

found casually by the inhabitants after e.g. a bout of rain with subsequent erosions, or picked up by anthropologists in a gathering of fossils brought by local collectors. Thus, such is the frequent case that the rough stratigraphic provenance or even the site of the find is unknown.

It is of paramount importance to develop a chronological framework in order to understand the significance of fossil finds. In the case of Sangiran hominids, locating the original source horizons of them is fundamental to the construction of such a framework. Analyses of selected elements such as fluorine in phosphatic skeletal remains sometimes provide a clue for allowing this allocation problem to be solved.

From 1977 to 1979, fieldwork on stratigraphy, palaeontology and chronology of human fossil bearing formations in Java, was conducted under the Indonesia-Japan Research Cooperation Programme CTA-41. Much attention was devoted to the Sangiran area, where the survey included diggings designed to observe the succession in detail at the fresh sections exposed and to collect vertebrate fossils *in situ*.

This paper deals primarily with the stratigraphic positions of *Pithecanthropus* and *Meganthropus* specimens from Sangiran as examined by the fluorine dating method using the excavated bone material as a guide series. This also gives some remarks about the possible significance of the results for the chronological positions of Sangiran hominids in human evolution.

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I Geological Structure, Stratigraphy and Fauna of Sangiran

Sangiran Village, about 12 km north of Surakarta, Central Java (Fig. 1), is located in the centre of a domelike uplift which is 4 km wide and 6 km long, and rises around 150 m above sea-level. Folding and erosion have exposed the late Cenozoic rock group in the Sangiran dome area (VON KOENIGSWALD, 1940; and others), producing

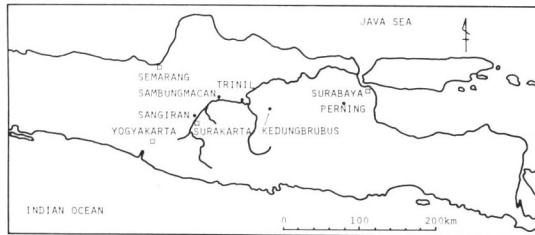


Fig. 1. Locations of Sangiran and some other sites of early man (small dots) in Java.

the structure that the oldest formation crops out in the central part encircled with younger layers in serial order (Fig. 2). However, the relative stratigraphic provenance of a spot can not be read merely from the distance from the centre of the dome to there; since the mid-dome area has a complicated geological structure due to faulting, depression and eruption of mud volcanoes (Fig. 3), and dissection by tributaries of the Solo River makes the outcrops along their ravines sometimes stretch over two or more formations.

The main part of the sequence of strata in the Sangiran dome area is subdivided into the Kalibeng, Pucangan, Kabuh and Notopuro formations in ascending order (Fig. 4). The Kalibeng formation has been referred to Pliocene, and the rest, Lower to Middle Pleistocene (Indonesia-Japan Joint Research Team, 1979). Late Pleistocene terrace deposits unconformably overlie the Kabuh and the Notopuro formation in

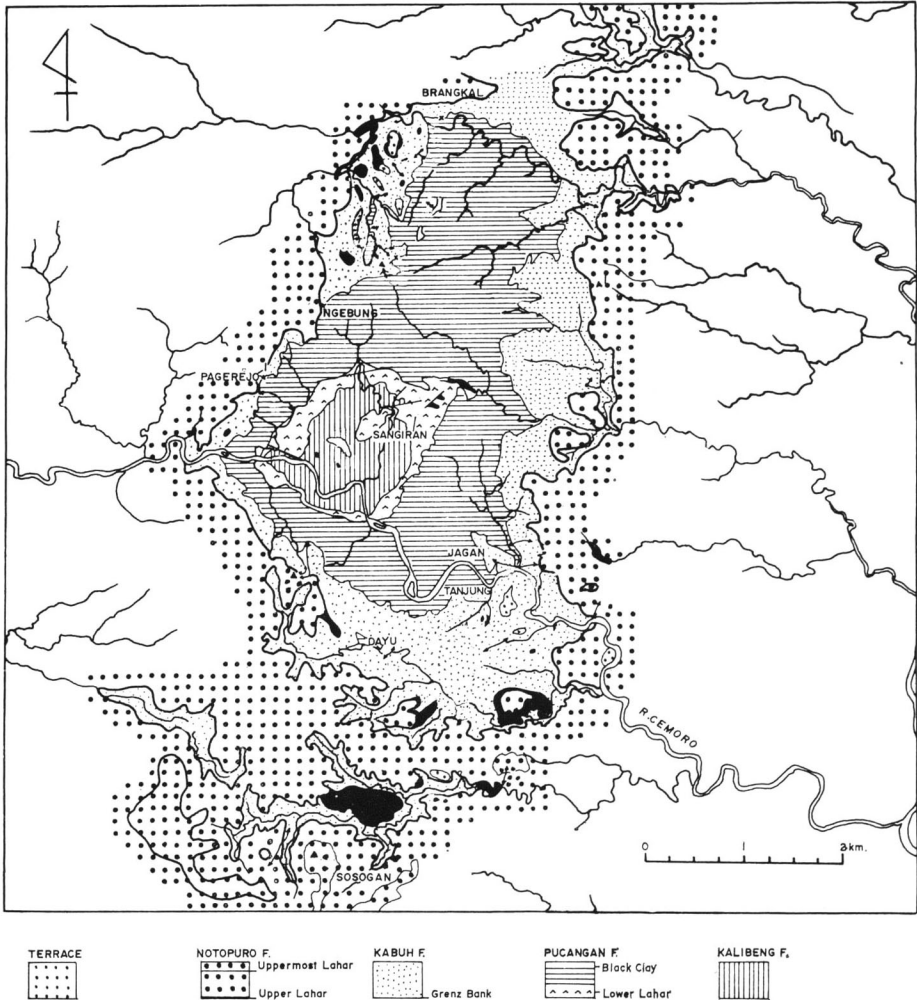


Fig. 2. Geological map of the Sangiran area (after Indonesia-Japan Research Cooperation Programme—CTA-41, 1979). See Fig. 3 for a detailed (revised) geological map of the central part of Sangiran Dome.

places, and Holocene alluvial deposits are distributed along rivers (Indonesia-Japan Research Cooperation Programme—CTA-41, 1979). The lithofacies and depositional environments of these sediments are summarized in Fig. 4 (see also Fig. 6).

The Pucangan and Kabuh formations are reported to contain the mammalian associations of the Jetis and Trinil faunae, respectively; Trinil elements also occur in the Notopuro formation of Sangiran, whose fauna is indistinguishable from that

of the Kabuh (VON KOENIGSWALD, 1940; VAN HEEKEREN, 1972). Note that VON KOENIGSWALD (1968a, p. 100) commented that it had not been confirmed whether the Grenzbank (see Fig. 4) 'should be regarded as the top of the Djetis [former spelling for Jetis] or the very base of the Trinil Beds'. Palaeontologists and anthropologists have frequently used the term, e.g. the Jetis layers (or beds) to represent that specific

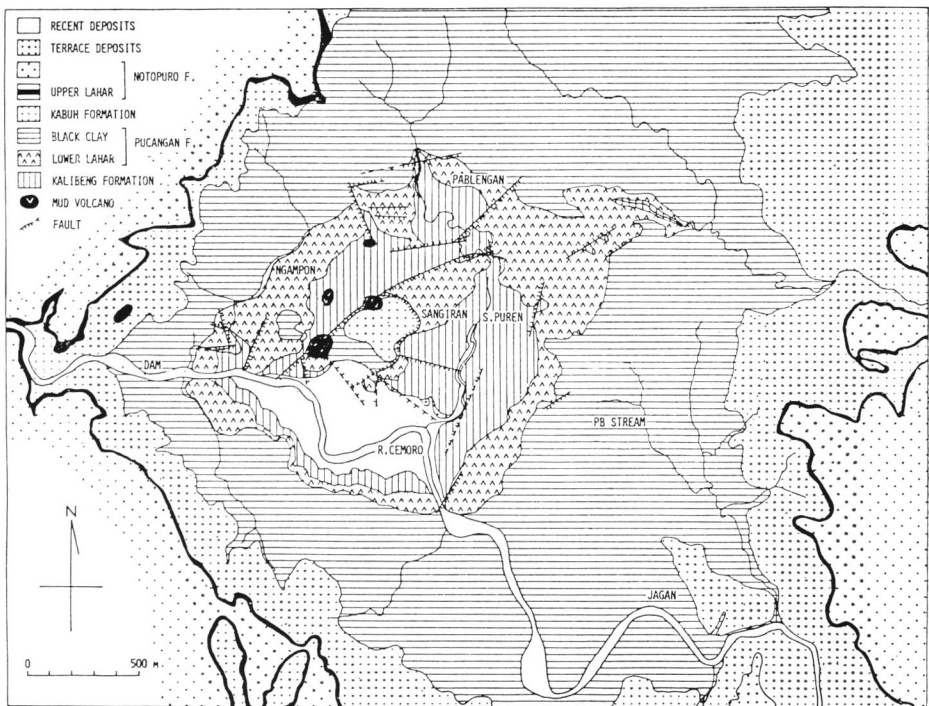


Fig. 3. Geological map of the central part of Sangiran Dome (after Indonesia-Japan Joint Research Team, 1979).

horizons in formation which yield guide fossils of the Jetis fauna. However, a recent palaeontological study in Java (AIMI & AZIZ, 1983) has proved that *Cervus lydekkeri*, guide fossil of the Trinil fauna, and *C. zwaani*, that of the Jetis fauna, occur *in situ* side by side in both the Kabuh and Pucangan formations of Sangiran Dome; and has further shown that the division of the Pleistocene faunae of Java is not so marked as previously described. Thus, time correlations of strata between the Sangiran area and other localities in Java according to the 'established' Javanese vertebrate stratigraphy would best be dismissed at present.

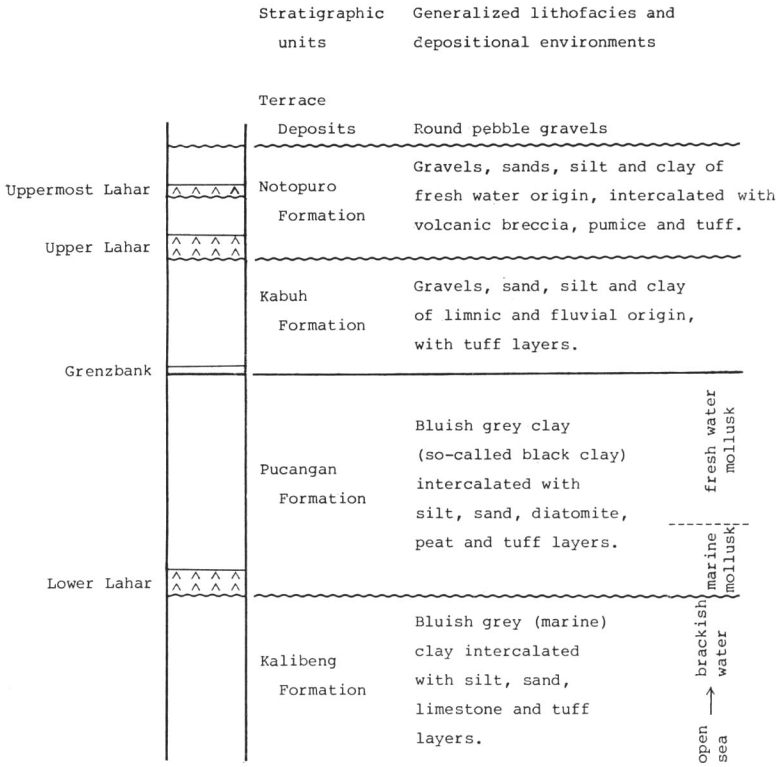


Fig. 4. Stratigraphic column of the Sangiran area. The total thickness of the formations is more than 380 m. Along rivers, recent alluvial deposits are also seen. Lahar, volcanic breccia; Grenzbank, calcareous sandstone layer with granule and pebble gravel. After Indonesia-Japan Research Cooperation Programme—CTA-41 (1979) and Indonesia-Japan Joint Research Team (1979).

II Fossil Hominid Remains of Sangiran

Discoveries of human fossils in Java, especially those after the World War II, have centred on the Sangiran area. Table 1 lists the Sangiran remains having code numbers as of 1980 (for reported spots of the finds, see Fig. 5). Remains of early man recovered from Java outside Sangiran include the juvenile calvaria of *Homo modjokertensis* from Perning (VON KOENIGSWALD, 1936: synonyms; *Pithecanthropus modjokertensis*, VON KOENIGSWALD, 1950; *Pithecanthropus* V, SARTONO, 1974, 1975), *Pithecanthropus* Mandible A from Kedungbrubus, *Pithecanthropus* Skull I from Trinil and an adult calotte from Sambungmacan (JACOB, 1975a).

Some specimens of Indonesian hominids are given registered designations in rough order of finding year mostly by the first publishers; viz., *Pithecanthropus* I, II, . . . , VIII for skull (excluding mandible) specimens with roman numerals annexed, and

Pithecanthropus A, B, . . . , F and Meganthropus A, B for mandible (maxilla in part) remains. Besides these designations a multiplicity of specific terms or nomenclature has been assigned to the early-hominid materials from Java (see Table 1), much confusing the problem of their taxonomic status.

LOVEJOY (1970) and CLARK (1978, and others) express the view that all of the Sangiran hominids including *Meganthropus* should be placed into a single species, *Homo erectus*. These lumpers, however, use the code or year of find to refer to an individual specimen. VAN HEEKEREN (1972) applies *Homo erectus erectus* to the *Pithecanthropus* fossils which were reported to be derived from the Kabuh formation, and *H. e. robustus* to those from the Pucangan formation; still, he separates individuals by putting marks, for instance, *H. e. e. II* and *H. e. r. C*.

Much attention should be rather paid where we are concerned with papers written or read by splitters who sometimes fail to quote a code of the specimen involved. For instance, 'the mandible of *Pithecanthropus modjokertensis* (VON KOENIGSWALD, 1973, p. 489)' implies Pithecanthropus B, since *P. (Homo) modjokertensis* of von Koenigswald at that time was represented by Pithecanthropus B and IV (see Table 1), and a number of isolated teeth as well as the holotype of *modjokertensis* from Perning.

In this paper I will employ such well-known designations as Pithecanthropus II, B; or Sangiran numbering (British Museum's Catalogue No.) for specimens without special designation. When confusion is apprehended, a note, e.g. 'holotype', will be added to the description of the fossil in order to facilitate identification.

Concerning cultural remains, stone artefacts have not yet been found in the hominid fossil bearing formations in Sangiran (BARTSTRA, 1982).

III Fluorine Dating Method

A. Historical Review

The fluorine dating method has a history approaching two centuries. The first observation of fluorine in fossilized bone material was made by MORICHINI in 1802 according to CARNOT (1893). As early as 1806, FOURCROY & VANQUELIN in France gave evidence by using both fossil and recent ivories that bone substances incorporate with 'fluoric acid' in course of burial in earth (see OAKLEY, 1951, 1953).

MIDDLETON (1844) first attempted to date bones by fluorine analysis; he inferred, for example, a fossil ruminant remain from the Sewalik (now written as Siwalik) Hills to be 7,700 years old on the hypothesis that the ratio fluoride/phosphate in fossil bone should be directly proportional to its antiquity. Some fifty years later a series of works by CARNOT (1892, 1893, and others) showed that the *average* content of fluorine in bone increases with geological age. On this ground VAN BEMMELEN (1897) made a fluorine test on a tibia of *Stegodon* from the layer that yielded Pithecanthropus Skull I to check the alleged age of the skull.

Carnot's research whereas proving the high potentialities of the fluorine-analysis approach, also drew attention to the fact that the fluorine content is so susceptible to

Table 1. Fossil hominid finds from Sangiran.

Code no.*	Material	Year of discovery	Stratigraphy**	Designation	Synonyms, classification & assignment (excluding <i>Pithecanthropus/Homo erectus/erectus erectus</i>)
S 1a	Rt. maxilla, M1-M3	?	?		<i>Meganthropus</i> (VON KOENIGSWALD in DAY, 1977)
S 1b	Rt. mandible, P4-M3	1936	Pucangan	Pithecanthropus B	<i>Pithecanthropus modjokertensis</i> (VON KOENIGSWALD, 1950)
S 2	Calotte	1937	Kabuh	Pithecanthropus II	
S 3	Parieto-occipital fr.	1938	Kabuh	Pithecanthropus III	
S 4	Calvaria, maxilla, It. C-M1, rt. C-M3	1938-39	Pucangan	Pithecanthropus IV	<i>Pithecanthropus robustus</i> , holotype (WEIDENREICH, 1945) <i>Pithecanthropus modjokertensis</i> (VON KOENIGSWALD, 1950)
S 5	Rt. mandible, M1, M2	1939	Pucangan		<i>Meganthropus</i> (VON KOENIGSWALD in WEIDENREICH, 1945) hominid or anthropoid? (WEIDENREICH, 1945)
S 6	Rt. mandible, P3-M1	1941	Pucangan	Meganthropus A	<i>Pithecanthropus dubius</i> , holotype (VON KOENIGSWALD, 1950) <i>Meganthropus palaeojavanicus</i> , holotype (VON KOENIGSWALD, 1950)
	Lt. mandible, M2, M3 ^{††}	1936	Pucangan	Meganthropus A	<i>Paranthropus palaeojavanicus</i> (ROBINSON, 1954) Meganthropus II (ROBINSON, 1953)
	Teeth	1937-41	P./K.		Meganthropus I (TOBIAS & VON KOENIGSWALD, 1964) Meganthropus II (TOBIAS & VON KOENIGSWALD, 1964)
S 7a/b	Mandible, It. M1-rt. M3 (only rt. M3 intact)	1952	Kabuh	Meganthropus B	<i>Meganthropus palaeojavanicus</i> (MARKS, 1953)
S 9	Rt. mandible, C-P4, M2, M3	1960	Pucangan	Pithecanthropus C	<i>Pithecanthropus dubius</i> (VON KOENIGSWALD, 1968a)
S 10	Calotte, It. zygoma	1963	Kabuh	Pithecanthropus VI	PV, sixth skull (JACOB, 1975a, p. 247)
S 11	Lt. upper M3, rt. lower II	1963	Kabuh?		
S 12	Calotte	1965	Kabuh	Pithecanthropus VII	
S 13a	Parieto-occipital fr.	1964	Kabuh		
S 13b	Calotte fragments	1965	?		
S 14	Cranial base fragment	1966	Kabuh		<i>Pithecanthropus soloensis</i> (JACOB, 1978)
S 15a	Lt. maxilla, P3, P4	1969	Pucangan?†		<i>Pithecanthropus modjokertensis</i> (SARTONO, 1974)
S 15b	Rt. maxilla, P3, roots of P4	1969	Kabuh		

Table 1. Continued.

Code no.*	Material	Year of discovery	Stratigraphy**	Designation	Synonyms, classification & assignment (excluding <i>Pithecanthropus/Homo erectus/erectus erectus</i>)
S 16	Rt. upper M2	1969	?		<i>Meganthropus</i> (VON KOENIGSWALD in JACOB, 1973) <i>Pongo</i> (JACOB, 1973)
	Lt. upper P3	1970	?		
S 17	Cranium, lt. P3, rt. C, M1-M3	1969	Kabuh	Pithecanthropus VIII	<i>Pithecanthropus soloensis</i> (JACOB, 1975b)
S 18a	Calotte fragments	1970	Kabuh		
S 18b	Calvarial fragments	1970	Kabuh		
S 19	Occipital	1970	Kabuh		
S 20	Calvarial fragments	1970/71	Kabuh		
S 21	Mandible, M3	1973	Kabuh	Pithecanthropus E	
S 22	Mandible, lt. I2-M1, rt. P4-M2; isolated teeth	1974	Pucangan	Pithecanthropus F	<i>Homo erectus trinilensis</i> (SARTONO, 1978) ⁺⁺
S 23	Endocast	1975	Kabuh		
S 24 [#]					
S 26	Cranial fragment [#]	1978	Pucangan		<i>Pithecanthropus modjokertensis</i> (JACOB, 1980)
S 27	Cranium	1978	Pucangan		<i>Meganthropus</i> , tentatively identified (JACOB, 1980)
S 34	Endocast, temporal fr.	1980	Kabuh		

* British Museum's Catalogue No.; S=Sangiran. B.M. Catalogue of Fossil Hominids—Part III (OAKLEY *et al.*, 1975) includes S 1 to S 22.

** All hominid remains from Sangiran are surface finds and in most cases is the exact finding site unknown. Thus the stratigraphic provenances are those inferred, presumed or alleged.

† WEIDENREICH later accepted the 1939 mandible as referable to a hominid (see LOVEJOY, 1970).

†† DAY (1977, p. 301) maintains that the left mandibular fragment does not appear to belong to the specimen of 1941 mandible. Actually, VON KOENIGSWALD (1969, p. 93) has regarded the 1936 find as the same *type* as the 1941 specimen.

+ B.M. Catalogue (OAKLEY *et al.*, 1975) lists S 15a in Kabuh remains. SARTONO (1975, p. 348), however, mentioned that "it may have come from the upper levels of the Early Pleistocene Putjangan beds." In this connection, see also SARTONO (1974).

++ SARTONO (see SARTONO *et al.*, 1981) proposes a new taxonomic scheme for Javanese Pleistocene hominids.

Briefly referred to by JACOB (1980).

A further specimen, a second lower deciduous molar of *Meganthropus*, is mentioned by VON KOENIGSWALD (1981).

burial environments that it may not be possible to estimate the absolute age of a bone merely from its fluorine measurement: and little cognizance appears to have been taken of the fluorine dating method in those days. This method had to await appreciation and revision by OAKLEY (1948) to find practical and great use for chronology of fossil bones. OAKLEY (1951, 1953, 1969, and others) has reintroduced the method especially to palaeoanthropology mainly in the following ways:

- 1) Where a human skeletal remain happens to have been eroded out of a deposit, or when there is a possibility of a hominid fossil, even one found from a particular bed, being derived from a lower layer or being intruded (through interment) from an upper layer; comparison of its fluorine content with that of a series of animal bones (so-called pilot series) from known horizons at the same or neighbouring site sometimes allows an inference for the original stratigraphic provenance of the specimen in question, and further for its relative geological age provided that an index fossil is found in the bones used for analytical comparison.
- 2) When bones (including a hominid fossil) of more than one age occur mixed in a single deposit through drifting, reaccumulation, etc.; the fluorine analysis may assist in differentiating them and in relating the human specimen to a particular fauna.

The fluorine dating method has been applied with varying degrees of success to many prehistoric human remains; for example, the *Eoanthropus* material, the most notable one (OAKLEY & HOSKINS, 1950; OAKLEY, 1951), the Galley Hill skeleton (OAKLEY & MONTAGU, 1949), the Swanscombe skull (OAKLEY, 1951, 1954), the Rhünda skull (OAKLEY, 1958), the Lagow skeleton (OAKLEY & HOWELLS, 1961), *Pithecanthropus* skull and femora from Trinil and the Wajak skulls (BERGMAN & KARSTEN, 1952), skeletal fragments from Mikkabi, Hamakita and Ushikawa (TANABE, 1962, 1966; TANABE & WATANABE, 1968), the Minatogawa man (MATSU'URA, 1982), etc.

Among the instances where comparison of fluorine content between hominid fossils and related fauna(s) has yielded no chronological information of use, we find the case of the Kanam jaw and the Kanjera skulls (WALLS in OAKLEY, 1951), and that of Amud man (TANABE, 1970). The former case of ill success was ascribed to profound mineral alteration (calcification) and rapid fluorine exchange which bones had undergone due to tropical weathering conditions and volcanic soils rich in fluoride; while the latter, to the arid environment that inhibits the introduction of fluorine to buried bones. The influences which environmental factors have on the rate of fluorine uptake by bone substance have been discussed by COOK (1960), GLOVER & PHILIPS (1965), PARKER & TOOTS (1976), SHIMODA (e.g. 1977) and others.

Whereas fluorine data on fossil hominid material from Pliocene and Pleistocene have made a great increase with assays (accompanied by phosphate and nitrogen determinations, and uranium estimation) that were involved in the publications of the "Catalogue of Fossil Hominids (OAKLEY *et al.*, 1971, 1975, 1977)", those on the associated faunal assemblages as comparative series are still scanty. Taking the case of the

Sangiran area, we can find in the Catalogue (Part III) only two fluorine analyses on animal fossils as compared with six ones on hominids. Thus it is of essential want to augment systematic collections of data for comparison.

B. Basics

Phosphatic skeletal material consists mainly of collagenous fibers and apatite crystals. Buried bones and teeth undergo diagenetic chemical changes. The protein collagen becomes less and less abundant during fossilization through hydrolysis reaction with subsequent leaching of amino acids by percolating water. The amount of protein remnant in a fossil is generally measured as nitrogen. Whereas the nitrogen concentration is about 5% in fresh bone (PROTSCH, 1975; HADDY & HANSON, 1982; and others), it rarely exceeds 0.1% in Early or Middle Pleistocene bones from middle latitudes (see WEINER *et al.*, 1955; OAKLEY *et al.*, 1971, 1975; KESSELS & DUNGWORTH, 1980; and others).

The inorganic phase, while receiving precipitation of mineral matter, probably of apatite in the main, is altered in composition after burial. Various elements present in surrounding soils and ground water are adsorbed in bone apatite and some of them are fixed in the crystal structure through ion-exchanges (NEUMAN & NEUMAN, 1958; PARKER & TOOTS, 1970).

The bone apatite, although it contains a minor quantity of carbonate (around 4% as CO₂: see e.g. BROPHY & NASH, 1968), is represented by:



The carbonate, chiefly in the form of CO₃OH³⁻, is likely to replace the PO₄³⁻ ions (see LEGEROS *et al.*, 1967; BROPHY & NASH, 1968; MATSUBARA, 1980).

Fluorine accumulates in buried bone (or teeth) at the cost of hydroxyl ions in the hydroxyapatite. This conversion proceeds in consequence of the higher molecular stability of fluorapatite over hydroxyapatite even under environmental conditions in low levels of fluoride (HAGEN, 1973; TSE *et al.*, 1973), approaching to the maximum theoretical content value of 3.77 wt. % F. The rate of the substitution reaction is affected by a series of factors, such as temperature, pH, the circumambient fluoride concentration, species of coexisting ions and the ionic strength (HAGEN, 1973).

The accumulation of fluorine in skeletal remains depends not only on the lapse of time and the above mentioned local circumstances, but presumably on the thickness and the physical properties (e.g. size of the apatite crystals, compactness or permeability) of the tissue that are to control the rate of fluoride diffusion into the fossil. The apatites of bone and dentine resemble in size and are both submicroscopic (PARKER & TOOTS, 1980). In contrast, enamel apatite crystals are estimated to average over 200 times larger than those of dentine, and are also much more dense (BRUDEVOLD & SÖREMARK, 1967). Therefore enamel is best separated from other phosphatic materials for comparison. *Substantia spongiosa* is liable to introduction of silt or other adventitious mineral grains. OAKLEY (1963; and others) has stated that this contamination problem can be eliminated by taking the ratio of F to P₂O₅ (a convenient measure

of the apatite content) in the sample.

The aforementioned bears that the fluorine dating needs due consideration of comparability in the consistency, texture type and burial context of the analysed samples.

To just locate the original source horizon of a hominid fossil demands that bones of different horizons be distinguished by fluorine determinations, not necessarily that the older show more fluorine than the younger. However, the content of this element in bones preserved in similar matrix at the same site or region in temperate climates, is known to increase gradually with age, thus providing a criterion for the relative antiquity of fossil bones (OAKLEY, 1951; GLOVER & PHILLIPS, 1965; MATSU'URA, 1978; and others).

IV Materials

A. Vertebrate Fossils

An abundance of vertebrate bones were collected *in situ* from human fossil bearing formations in Sangiran by the CTA-41 research team (1977-79). Figs. 5 and 6 show the excavation sites and the stratigraphical contexts of those sites, respectively. Also indicated in Fig. 6 are the horizons of the vertebrate fossil zones, ranging between the Upper Tuff (Kabuh f.) and Tuff 3 (Pucangan f.). Such a systematic collection of fossil material would be the first to come by. No mammalian fossil beds have been detected by the fieldwork of the CTA-41 project in the strata above the Upper Tuff in the Kabuh formation nor below Tuff 3 in the Pucangan.

The facies of the vertebrate beds involved (see Fig. 6) are:

Kabuh Formation

- 1) between the Upper and the Middle Tuff—yellow to yellowish brown, sand and gravel with pumice ball;
- 2) between the Middle and the Lower Tuff—yellowish white, sand with granule and pebble;
- 3) just below the Lower Tuff—yellowish white, sand with pebble;
- 4) Grenzbank, the basal part—brownish yellow, sand and sandstone with granule and pebble conglomerates;

Pucangan Formation

- 1) above Tuff 11 (Cengklik site)—bluish grey clay;
- 2) above Tuff 11 (Jagan site)—black to dark bluish grey, silt and clay;
- 3) between Tuff 10 and Tuff 9—dark grey to brownish grey, silt and clay;
- 4) just below Tuff 5—brownish grey to black, clay;
- 5) between Tuff 4 and Tuff 3—brownish grey silty clay.

Two hundred and thirty five specimens of the excavated bones were used in this study. Twelve specimens unearthed *in situ* around the sites of *Pithecanthropus* II, VII and VIII also formed the material. Besides, several surface finds of fossilized animal remains from the Kabuh and Pucangan formations were employed for pre-

liminary examinations.

A list of the faunal assemblage collected by the CTA-41 team is available in AIMI & AZIZ (1983).

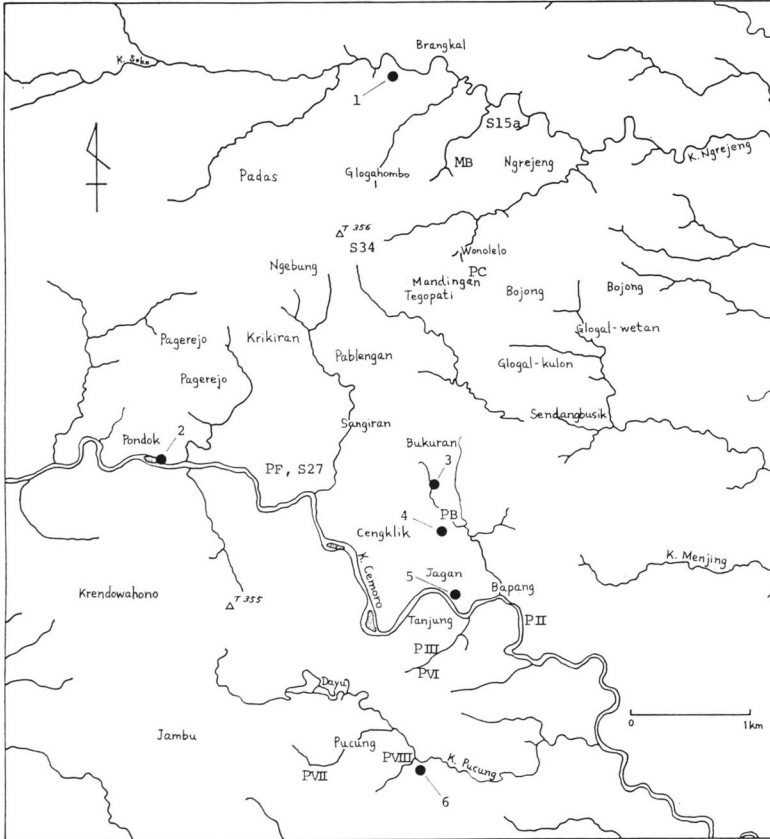


Fig. 5. Map of the Sangiran area, showing reported localities of hominid fossils (P= Pithecanthropus, M=Meganthropus, S=Sangiran; see Table 1), and pointing the excavation sites (solid circle) of vertebrate fossils which formed the material for most of this study. Exc. site name: 1, Brangkal; 2, Pondok; 3, Bukuran; 4, Cengklik; 5, Jagan; 6, Pucung.

B. Pithecanthropus VII and VIII

Small chips belonging to the Pithecanthropus VII and VIII skulls were provided by the Geological Research & Development Centre in Bandung through the courtesy of Dr. Darwin KADAR, Chief of the Palaeontological Laboratory of the same institution.

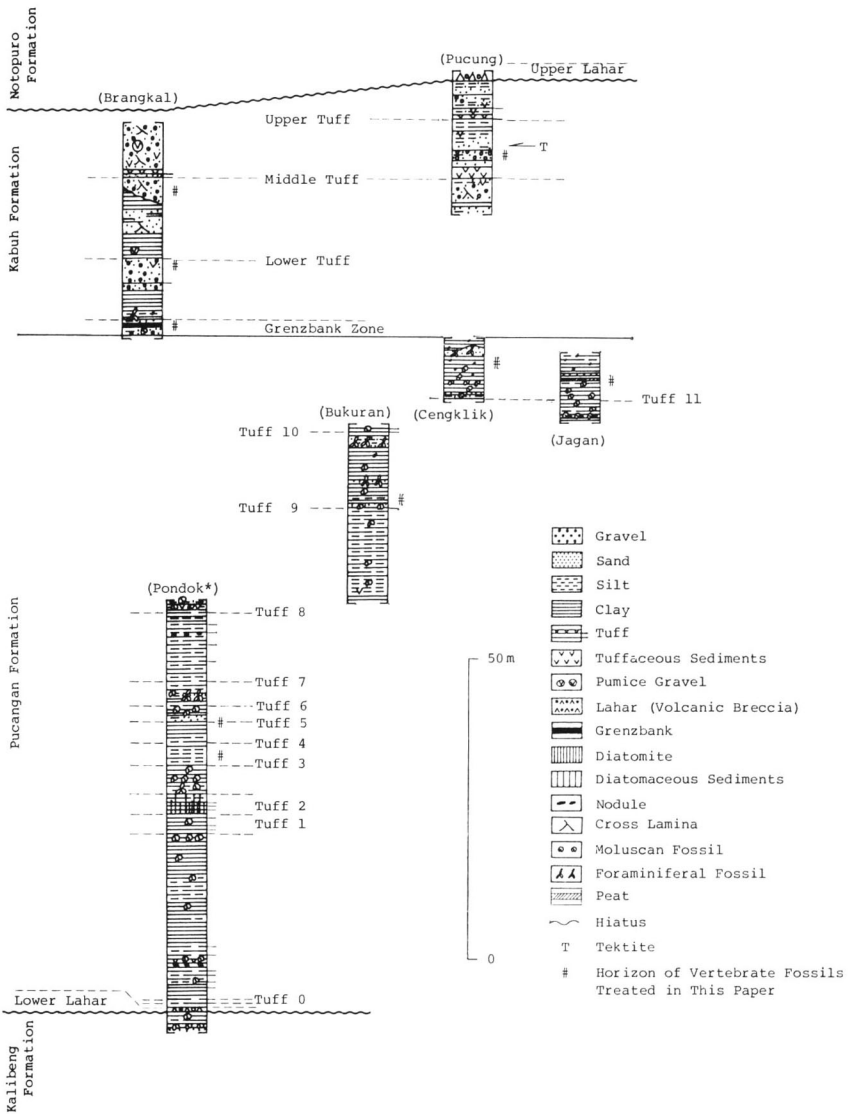


Fig. 6. Geological columnar sections of the Kabuh and Pucangan formations developed at the six excavation sites of the CTA-41 project (after ITHARA, 1981, personal communication). The Kabuh formation is tentatively subdivided into four: the lowermost part, from Grenzbank to the base of the Lower Tuff; lower part, from the Lower Tuff to the base of the Middle Tuff; middle part, from the Middle Tuff to the base of the Upper Tuff; upper part, from the Upper Tuff to the base of the Upper Lahar (IJRCP—CTA-41, 1979).

* Compiled with the columnar section made around the Pablengan village for the part below Tuff 2.

V Analytical Methods

A. Sample Preparation

The vertebrate fossils were rinsed in tap water, and dried in the sunlight.

1. *Substantia Compacta* of Bone and Antler.

A small transverse section extending from the outer to the inner surface of compact tissue was cut from each fossil by means of a fret saw or a dental machine equipped with a diamond disc blade. After surface contaminants were removed using a stiff nylon brush, the sample was ground with an agate mortar to a fine and homogeneous powder. Ultrasonic cleaning in deionized water and natural drying prior to grinding were employed if required.

Where partial zonal analysis of fluorine in *substantia compacta* was to be carried out, the requisite amount of powder was obtained by drilling a hole while measuring the depth from the outer (or inner) surface, and further pulverized in a mortar.

The chips of *Pithecanthropus* VII and VIII were powdered without any treatment.

2. Tooth.

A piece of cross section was taken out of dentine and enamel separately. Where the different tooth materials were difficult to separate, the dentine part was removed with a dentist's drill. The samples were pulverized to be fine.

3. *Substantia Spongiosa*.

Samples were obtained using a dental drill and well powdered without any particular treatment even in the case of a specimen contaminated by soil or other foreign minerals.

B. Chemical Analyses

All chemicals were of guaranteed reagent grade or super special grade, and water distilled from an all-glass still was used throughout.

1. Fluorine.

Fluorine was determined electrometrically using the fluoride ion-sensitive membrane electrode (FRANT & ROSS, 1966; EVANS *et al.*, 1971; MOODY & THOMAS, 1971). MATSU'URA (1981) discussed and described procedures for fluorine determination of fossil bone with a fluoride electrode, on which the following method is based.

About 10 mg of the bone powder is weighed to an accuracy of 0.05 mg and dissolved in 5 ml 1 M HCl. After 5 ml water is added, the total ionic strength and pH of the bone solution are fixed by adding 30 ml of a buffer which is prepared according to TISAB IV (see Orion Inc., 1977) with modulation for the assay of bones (MATSU'URA, 1981). The quantities may be scaled down or up as appropriate. Final concentrations of the buffer medium are 0.5 M HCl, 1 M TRIS and 0.5 M NaTartrate.

The fluoride electrode and a reference electrode (S.C.E.) are placed in the solution which is kept stirred with a Teflon-coated magnetic bar. The potential is recorded

after an 20 min equilibrium period. Fluorine concentration can be determined by reference to a calibration curve constructed with several NaF standards of the same ionic strength and pH as the sample solution.

Repeated weight measurements and subsequent fluorine determinations by the direct measurement procedure described above on pulverized bone samples gave a coefficient of variation (standard deviation/mean), about 0.012, showing a high degree of reproducibility of this method. The direct measurement results were verified by known addition procedure.

In assay of soil specimens as will be mentioned in Chapter VI A-5, HCl-NaCitrate (pH 5.5-6.5) and Denki Kagaku Keiki's TISAB (DKK, 1976) media were employed since the fluoride content in soil extracts was not high enough to be measured with high precision using the HCl-TRIS-NaTartrate buffer system (see MATSU'URA, 1981).

2. Phosphate.

Phosphate was determined by acid-base titration after precipitation as ammonium phosphomolybdate (see e.g. TAKAGI, 1967, 1969; TIBA, 1970).

Five to 10 mg of prepared bone sample (in case of spongy tissue, approximately 15 mg) is weighed accurately and dissolved in a 50-ml beaker by addition of 10 ml of warm 2.64 M HNO₃ (61% nitric acid diluted 1+4), and then filtered through a TOYO No. 5B paper into a 200-ml beaker. The 50-ml beaker is washed several times with 25 ml warm 2.64 M NHO₃ and 20 ml water in total, which are collected through the filter in the 200-ml beaker. After addition of 20 ml of NH₄NO₃ solution (340 g/l), the filtrate is heated to 40-45°C, then mixed with 16 ml of 3% ammonium molybdate solution, and allowed to stand at least overnight at room temperature. The reaction mixture is filtered through a TOYO No. 5C paper and the filtrate is discarded. The precipitate is sequentially washed with three 20-ml portions of a solution containing 50 g NH₄NO₃ and 20 ml conc. HNO₃ per liter, three 20-ml portions of 1% HNO₃ solution, and finally 1% KNO₃ until the washings become neutral. The filter paper retaining the precipitate is transferred to the 200-ml beaker and demolished in water. A specific volume of 0.1N NaOH is added to dissolve the precipitate. An excess of NaOH is titrated with 0.1N HNO₃ with phenolphthalein used as the indicator. Phosphate content can be found from the volume of the NaOH solution consumed to dissolve the precipitate using:

$$1 \text{ ml of } 0.1N \text{ NaOH} \equiv 0.309 \text{ mg P}_2\text{O}_5$$

The reproducibility was $\pm 3\%$ (range) of the result.

VI Standard Data for the Fluorine Dating of the Sangiran Hominids

Fossil remains of hard tissue of vertebrates involve bone, antler (or horn core), tortoise shell and tooth. Antler, tortoise shell and cementum of tooth are specialized

or particular types of bone. Dentine is similar to bone in both physical and chemical properties, and thus used to be grouped together with bone in the fluorine dating method. Enamel, in contrast, should best be dealt with separately from the other bony material, as described in Chapter III, B.

Spongy tissue is liable to mineral alteration and what is more unfavorable, is difficult of complete removal of intrusive silts or foreign mineral matter. It would thus fail to form a material suitable for examining the difference of fluorine content of bone among horizons or for assessing the rate of fluorine accumulation. In the case of human fossils, however, frequently only samples of cancellous or spongy regions can be spared for chemical test, and it is of practical importance to investigate on that kind of tissue. In this paper the fluorine concentration of *substantia spongiosa* will be discussed in the light of its correlation with that of *substantia compacta* from the same specimen (or fragment).

A. *Substantia Compacta* and Dentinum

1. Depth Dependent Variation of Fluorine Concentration in Compact Bone.

Table 2 gives the results of zonal analysis of fluorine in compact bone when trisected; Table 3, those when bisected. Such variation in fluorine content of fossil bone with depth from its exposed surfaces as shown in the tables has a concern with comparison of measured fluorine values between a hominid fossil and a pilot series of animal bones.

Data on animal specimens that may construct a comparative table for fluorine dating are obtained generally by analysing samples of cores or sections extending from the outer surface to the inner surface of compact tissue. On the other hand, it is usually the case with fossil hominid material that only a limited portion of the compact

Table 2. Zonal fluorine analysis of *substantia compacta*. A bone sample was zoned into three equal parts; outside, middle and inside.

Specimen	Formation	Thick- ness of compact tissue (mm)	Average F (%)	Outside		Middle		Inside		
				F (%)	R.D.*	F (%)	R.D.*	F (%)	R.D.*	
vertebrate	Kabuh	6.9	2.96	2.97	0	3.09	+4	2.81	-5	
mammal	Kabuh	8.3	2.50	2.50	0	2.47	-1	2.53	+1	
cervid, metapodial**	Pucangan	2.5	1.45	1.61	+11	1.42	-2	1.32	-9	
" "	" **	"	2.8	1.35	1.57	+16	1.28	-5	1.20	-11
" "	" **	"	3.2	1.35	1.55	+15	1.27	-6	1.23	-9
" "	" **	"	3.3	1.37	1.57	+15	1.35	-1	1.19	-13
mammal, long bone	Pucangan	4.5	1.15	1.23	+7	1.16	+1	1.07	-7	
mammal, pelvis	Pucangan***	5.8	0.824	0.918	+11	0.800	-3	0.755	-8	

* Zonal deviation relative (%) to average fluorine content.

** From the same bone.

*** Surface collection.

Table 3. Zonal fluorine analysis of compact bone and tusk. A sample was bisected into outer and inner zones.

Specimen	Formation	Thickness of compact tissue (mm)	F (%)			R.D.*
			Average	Outer Z.	Inner Z.	
vertebrate	Kabuh S**	6.0	2.80	2.75	2.85	± 2
vertebrate	Kabuh	4.1	2.76	2.82	2.69	± 2
vertebrate	Kabuh S	5.9	2.64	2.75	2.52	± 4
vertebrate	Kabuh	5.5	2.20	2.26	2.14	± 3
elephantid, tusk	Kabuh	>9	1.65	1.68	1.61	± 2
mammal, long bone	Pucangan S	3.9	1.58	1.60	1.55	± 2
cervid, metapodial***	Pucangan	2.3	1.45	1.62	1.28	±12
" " ***	"	2.2	1.43	1.55	1.30	± 9
mammal	Pucangan	3.3	0.947	0.953	0.940	± 1
mammal	Pucangan S	4.2	0.800	0.913	0.687	±14

* Zonal deviation relative (%) to average fluorine content.

** S= Surface collection.

*** From the same bone.

region is available for analysis. Hence, zonation of fluorine should receive deliberation.

Since diffusion is to be a factor of the accumulation of fluorine in bone (see Chapter III, B), this element is expected to be more uniformly distributed throughout the bone substance as it becomes closer to saturation (i.e. 3.77% F). This sort of trend is suggested by the data on the Sangiran specimens (Tables 2 & 3). In bones where a diffusion gradient is apparent, the deviation of fluorine content is larger at the outside portion (Table 2). Considering we can not suppose that a sufficiency for relative dating procedures is removed exclusively from the surface layer (expected in many cases to have the highest local concentration of fluorine) of a precious hominid fossil, it would be substantial in practice that we allow $\pm 15\%$ variability for a fluorine data on a fossil hominid obtained using a compact bone sample.

2. Intra-Specimen Variation in Fluorine Content of Cross Sections of Compact Bone.

Provided that fluorine is more concentrated in the surface layers of bones, thinner parts within a compact bone fragment may be expected to have a higher average fluorine content than that of thicker parts. Fig. 7 shows that this tendency is well illustrated by fossil bones with lower total fluorine; this is consistent with the fact that the concentration gradient from the outside to the inside of a bone tends to be evident in samples having lower fluorine as observed in Tables 2 & 3. Even in the case of bones with higher fluorine content, large and thick ones would show the tendency of relatively thinner parts having more fluorine on the average; this has been suggested by a fossil bone from the Kabuh formation (Fig. 7, upper right: mammal, Kabuh). Variation of fluorine content among compact bone samples of similar thickness taken

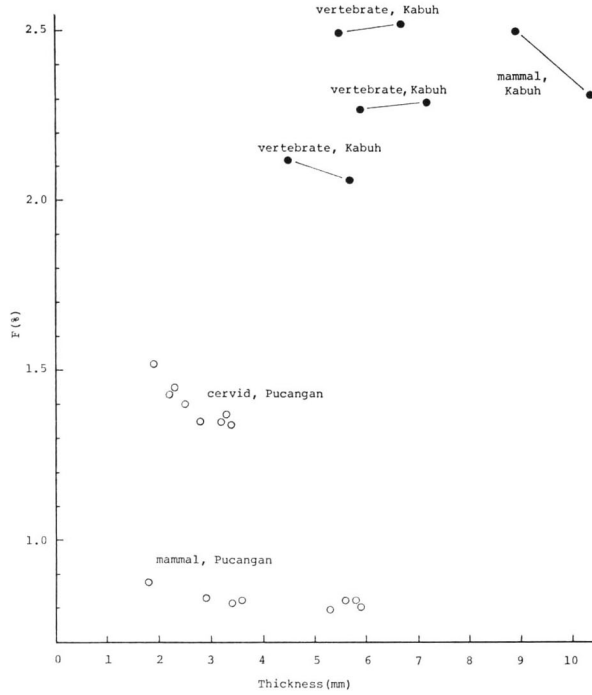


Fig. 7. Intra-specimen variation of fluorine content in *substantia compacta* with the thickness of the analysed part.

Table 4. Fluorine content of compact bone samples with about the same ($\pm 10\%$) thickness taken from a given specimen.

Specimen	Formation	T* (mm)	F (%)	T* (mm)	F (%)	T* (mm)	F (%)	Variation (%)
vertebrate	Kabuh	6.9	2.96	6.9	2.92			1
vertebrate	Kabuh	4.4	2.69	4.4	2.68			0
vertebrate	Kabuh	6.4	2.52	6.7	2.54			1
vertebrate	Kabuh S**	4.4	3.48	4.8	3.45			1
vertebrate	Kabuh	4.1	2.32	4.2	2.33			0
vertebrate	Kabuh	3.5	2.19	3.5	2.10			4
vertebrate	Kabuh	3.8	2.10	3.9	2.12			1
cervid, metapodial†	Pucangan	2.2	1.43	2.3	1.45			1
" "	"	2.3	1.45	2.5	1.40			4
" "	"	3.2	1.35	3.3	1.37	3.4	1.34	2
mammal, pelvis††	Pucangan S	3.4	0.816	3.6	0.824			1
" "	"	5.3	0.797	5.6	0.824	5.8	0.824	3
" "	"	5.6	0.824	5.8	0.824	5.9	0.803	3

* Thickness of compact tissue.

** S=Surface collection.

† From the same bone.

†† From the same bone.

from a given specimen, is small (not more than 4%) for the Sangiran specimens, no matter what the average content is (Table 4).

The above findings suggest that where a bone specimen varies wide in thickness, it is desirable to analyse fluorine at two or more parts of different thickness. Yet, this procedure would be in general unpractical because it may seriously damage the specimen. In this study, I constructed comparative fluorine data by analysing bones with varying thicknesses for each horizon in order to cover the probable variance in fluorine content dependent on thickness.

3. Fluorine Content of Vertebrate Fossils *in situ* from Various Horizons.

Tables 5–16 give the resulting data for fluorine analysis. Figs. 8–16 show the

Table 5. Analysis of fluorine in compact bone and dentine of fossil specimens *in situ* from the CTA-41's excavation site at Pucung.

Horizon: between the Upper and the Middle Tuff in the Kabuh formation; see Fig. 6.

Specimen	Thickness of analysed part* (mm)	F (%)	Specimen	Thickness of analysed part* (mm)	F (%)
mammal	10.5	2.31	mammal	6.8	2.66
"	10.4	2.36	vertebrate	6.9	2.68
"	4.3	2.39	mammal	2.0	2.69
"	5.1	2.42	"	7.1	2.76
"	4.1	2.49	"	2.1	2.76
"	8.3	2.50	vertebrate	2.9	2.78
" , tooth	3.5	2.55	mammal	5.3	2.80
mammal	6.2	2.56	"	1.8	2.80
vertebrate	2.1	2.61	"	6.1	2.82
mammal	2.7	2.61	"	2.1	2.87
"	4.5	2.63	"	2.6	2.98

* Thickness of compact tissue or dentine. (Common to Tables 5–16.)

Table 6. Analysis of fluorine in compact bone of fossil specimens collected *in situ* near the site of Pithecanthropus II.

Horizon: the Kabuh formation excluding Grenzbank; exact horizon not ascertained.

Specimen	Thickness of analysed part (mm)	F (%)
vertebrate	2.5	2.31
"	3.4	2.34
"	2.5	2.44
"	2.0	2.64
"	2.2	2.66

Table 7. Analysis of fluorine in compact bone of fossil specimens collected *in situ* near the site of Pithecanthropus VII.

Horizon: the Kabuh formation excluding Grenzbank; exact horizon not ascertained.

Specimen	Thickness of analysed part (mm)	F (%)
vertebrate	2.3	2.65
tortoise, shell	1.6	2.68
vertebrate	2.9	2.69
"	1.0	3.07

Table 8. Analysis of fluorine in compact bone and dentine of fossil specimens *in situ* from the CTA-41's excavation site at Brangkal.

Horizon: between the Middle and the Lower Tuff in the Kabuh formation; see Fig. 6.

Specimen	Thickness of analysed part (mm)	F (%)	Specimen	Thickness of analysed part (mm)	F (%)
vertebrate	8.5	2.39	vertebrate	1.9	2.79
"	5.4	2.63	"	5.6	2.79
"	3.6	2.65	"	7.6	2.80
"	4.2	2.65	"	2.3	2.81
"	2.7	2.66	"	6.3	2.82
"	7.4	2.66	elephantid, tooth	8.9	2.84
"	4.9	2.67	vertebrate	5.6	2.92
"	6.5	2.70	"	6.9	2.94
"	3.3	2.70	"	5.2	2.95
"	5.6	2.74	"	6.7	2.96
"	6.5	2.75	"	5.9	2.99
"	4.4	2.75	"	5.2	3.07
"	6.3	2.77	"	5.3	3.08
"	5.8	2.78	"	4.9	3.16

Table 9. Analysis of fluorine in compact bone of fossil specimens collected *in situ* at the site of Pithecanthropus VIII.

Horizon: probably between the Middle and the Lower Tuff in the Kabuh formation.

Specimen	Thickness of analysed part (mm)	F (%)
vertebrate, rib?	2.0	2.63
vertebrate	4.1	2.76
"	0.9	2.90

Table 10. Analysis of fluorine in compact bone and dentine of fossil specimens *in situ* from the CTA-41's excavation site at Brangkal.

Horizon: just below the Lower Tuff in the Kabuh formation; see Fig. 6.

Specimen	Thickness of analysed part (mm)	F (%)	Specimen	Thickness of analysed part (mm)	F (%)
vertebrate	4.2	2.33	vertebrate	1.8	2.85
"	8.3	2.45	"	3.7	2.85
" , tooth	6.1	2.46	"	1.7	2.86
vertebrate	5.1	2.54	"	6.4	2.88
"	5.1	2.59	"	2.4	2.89
"	1.8	2.62	"	2.1	2.90
" , tooth	2.9	2.64	"	5.6	2.91
vertebrate	4.6	2.65	"	2.9	2.92
"	1.5	2.74	"	5.4	2.93
"	6.4	2.79	"	2.7	2.97
"	6.9	2.83	"	3.2	3.03

Table 11. Analysis of fluorine in compact bone and dentine of fossil specimens *in situ* from the CTA-41's excavation site at Brangkal.
Horizon: Grenzbank, see Fig. 6.

Specimen	Thickness of analysed part (mm)	F (%)	Specimen	Thickness of analysed part (mm)	F (%)
elephantid, tusk	>9.0	1.65	vertebrate	5.2	2.12
vertebrate, tooth	7.4	1.69	"	5.6	2.13
vertebrate	3.2	1.70	"	6.3	2.14
"	7.5	1.81	"	3.5	2.14
"	3.9	1.82	"	4.3	2.16
"	7.0	1.85	"	2.6	2.16
"	6.8	1.87	"	8.2	2.18
elephantid, tusk	>9.3	1.90	"	4.3	2.18
vertebrate	8.0	1.91	"	4.4	2.18
"	2.5	1.95	"	4.2	2.19
"	8.3	1.95	"	5.5	2.20
"	3.4	1.97	"	7.8	2.20
"	2.2	1.98	"	4.5	2.25
"	3.9	1.98	"	5.0	2.26
"	2.6	1.99	"	6.2	2.27
"	5.0	2.02	"	5.9	2.28
"	3.7	2.03	"	7.2	2.33
"	7.8	2.04	"	7.1	2.34
"	3.6	2.04	"	9.1	2.35
"	6.9	2.06	"	7.3	2.39
"	4.1	2.07	"	7.8	2.39
"	9.0	2.09	"	5.5	2.50
"	3.8	2.09	"	4.0	2.52
"	5.7	2.09	"	6.5	2.53
"	4.7	2.10	"	6.7	2.57
"	3.3	2.10	"	3.8	2.58
"	6.6	2.10	"	4.4	2.69
"	7.8	2.11	"	5.0	2.76
"	3.9	2.11	"	2.4	2.77

relation between the fluorine content and thickness of compact tissue (or dentine) for skeletal remains excavated from 9 vertebrate fossil beds (see Chapter IV) respectively.

First, I refer to the analyses on dentine. The rate of fluorine uptake by dentine has been believed to be practically identical with that by compact tissue of bone (see e.g. OAKLEY, 1951). This seems to be supported by the data in Table 17. It will be noted, however, that the fluorine content of dentine samples from Sangiran tends to

Table 12. Analysis of fluorine in compact bone and horn of fossil specimens *in situ* from the CTA-41's excavation site at Cengklik.

Horizon: above Tuff 11 in the Pucangan formation; see Fig. 6.

Specimen	Thickness of analysed part (mm)	F (%)	Specimen	Thickness of analysed part (mm)	F (%)
tortoise, shell	2.2	1.12	mammal, rib	2.1	1.28
cervid, tibia	3.7	1.15	bovid, femur	4.3	1.29
<i>Bibos palaesondaicus</i> , maxilla	2.9	1.17	tortoise, shell	2.5	1.30
mammal, phalanx	4.4	1.18	" "	1.9	1.33
" , rib	7.3	1.21	" "	1.8	1.36
" , horn	1.9	1.21	" "	1.5	1.36
" , rib	3.4	1.25	bovid, radius	6.8	1.40
mammal	4.9	1.25	mammal, vertebra	1.4	1.41
bovid, mandible	5.6	1.26	" , rib	2.2	1.42
mammal, rib	2.6	1.27	" , vertebra	1.9	1.43
			" "	1.4	1.46

Table 13. Analysis of fluorine in compact bone and dentine of fossil specimens *in situ* from the CTA-41's excavation site at Jagan.

Horizon: above Tuff 11 in the Pucangan formation; see Fig. 6.

Specimen	Thickness of analysed part (mm)	F (%)	Specimen	Thickness of analysed part (mm)	F (%)
vertebrate, tooth	6.5	1.27	vertebrate	1.3	1.63
vertebrate	2.0	1.45	"	1.2	1.71
"	3.4	1.47	fish	0.8	1.78
tortoise, shell	1.3	1.56	vertebrate	0.9	1.81
mammal	1.1	1.56	"	0.8	1.92

Table 14. Analysis of fluorine in compact bone and antler and in dentine of fossil specimens *in situ* from the CTA-41's excavation site at Bukuran.

Horizon: between Tuff 10 and Tuff 9 in the Pucangan formation; see Fig. 6.

Specimen	Thickness of analysed part (mm)	F (%)	Specimen	Thickness of analysed part (mm)	F (%)
mammal	3.3	0.921	mammal, rib	1.9	1.19
cervid?, femur	3.1	0.936	mammal	4.5	1.24
bovid, humerus	6.1	0.962	"	4.0	1.29
tortoise, shell	6.7	0.968	"	1.5	1.34
mammal, rib	2.5	0.982	cervid, metapodial	2.8	1.35
mammal	3.4	0.984	<i>Cervus zwaani</i> , mandible	2.4	1.35
" , humerus	3.4	1.02	tortoise, shell	2.3	1.37
" , rib	3.8	1.02	cervid, antler	2.3	1.39
mammal	2.5	1.03	tortoise, shell	1.3	1.43
" , tooth	2.9	1.03	" "	1.2	1.47
" , rib	3.5	1.07	" "	2.7	1.48
<i>Cervus zwaani</i> , mandible	2.1	1.07	mammal	3.4	1.49
crocodile, tooth	5.1	1.11	fish	1.9	1.49
<i>Cervus zwaani</i> , mandible	2.2	1.13	mammal, vertebra	1.3	1.49
mammal, phalanx	1.7	1.15	mammal	2.6	1.53
cervid?, ulna	3.0	1.16	" , phalanx	0.8	1.62
mammal, rib	3.7	1.16	tortoise, shell	0.8	1.64
cervid, antler	2.5	1.19			

Table 15. Analysis of fluorine in compact bone and dentine of fossil specimens *in situ* from the CTA-41's excavation site at Pondok.

Horizon: just below Tuff 5 in the Pucangan formation; see Fig. 6.

Specimen	Thickness of analysed part (mm)	F (%)	Specimen	Thickness of analysed part (mm)	F (%)
bovid, tibia	5.0	0.917	mammal	4.0	0.977
mammal	5.1	0.928	"	5.1	0.982
"	6.8	0.931	"	3.5	1.01
<i>Cervus zwaani</i> , tooth	6.2	0.947	"	3.7	1.02
mammal	4.8	0.953	"	4.9	1.02
"	3.9	0.965	"	2.7	1.07
"	4.4	0.973	<i>Cervus zwaani</i> , mandible	1.7	1.11

Table 16. Analysis of fluorine in compact bone and antler and in dentine of fossil specimens *in situ* from the CTA-41's excavation site at Pondok.

Horizon: between Tuff 4 and Tuff 3 in the Pucangan formation; see Figs. 6.

Specimen	Thickness of analysed part (mm)	F (%)	Specimen	Thickness of analysed part (mm)	F (%)
vertebrate, tooth	14.0	0.821	mammal	3.3	0.947
mammal	5.4	0.865	"	2.4	0.956
<i>Cervus hippelaphus</i> ,			"	2.7	1.00
antler	3.2	0.893	"	1.6	1.05
mammal	6.7	0.898	"	3.5	1.08
"	7.1	0.906	cervid, antler	1.5	1.10
"	4.4	0.911	" "	2.1	1.11
"	4.2	0.916	<i>Cervus hippelaphus</i> ,		
"	2.0	0.932	antler	1.4	1.16
"	5.5	0.934	mammal	4.3	1.16
"	6.2	0.936	"	2.3	1.16
cervid, antler	2.0	0.936	cervid, antler	1.4	1.18
mammal	5.2	0.939	" , tooth	0.9	1.29
"	4.5	0.943			

be a little lower than that of compact bones with comparable thickness and from the same horizon, though the former content is likely to fall within the range of the latter (Figs. 8–11, 13–16). This point requires future investigations on a larger scale and with due consideration of the preservation of teeth.

Where we compare analyses of hominid specimens with those of animal bones, the differences in fluorine content attributable to taxon might be a matter in question.

In palaeontological material it is not possible to carry out experiments in the strict sense on this matter because we can not obtain fossil bones in which all variables are kept constant except for species. However, Figs. 12–14, where mammal bones and tortoise shells from each horizon are differentiated by two types of point symbols and where a systematic bias is not yet found between the fluorine concentrations of the two groups, should hint that taxon has a minor, if any, influence on the amount of this element accumulated in bone during fossilization.

Next as concerns the correlation between the fluorine content and thickness of bone, it was significant for 5 out of 9 horizons (Figs. 8–16). Data on dentine samples were excluded for the aforementioned reason from calculations of the correlation coefficients. The significant negative correlations are found mostly in sets of fossils (by the horizon) which show the relatively lower fluorine. This is to correspond to what has been observed in the next section ahead.

Factors which cause the fluctuation in fluorine content of bones derived from a given stratum may include the thickness and preservation of bone, the time span of the stratum, minor changes in facies, and differences in compactness of the deposit. Thickness of bone can well explain the variance of fluorine content of fossil bones from a horizon just below Tuff 5 in the Pucangan formation (Fig. 15; coefficient of determination, $r^2=0.70$); yet, the dispersions seen for the other 8 horizons, even where a significant correlation between thickness and fluorine content was found, are ascribable rather to the other factors. Of these factors “time span” is to be a minor one, since each fossil bed is estimated to span a few tens of thousand years, which is not

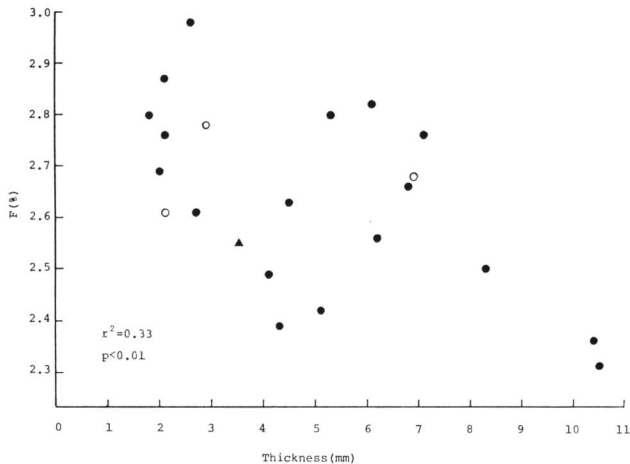


Fig. 8. Fluorine content of compact bone and dentine plotted against thickness of the specimen at the analysed part (exact figures given in Table 5). ○, vertebrate bone; ●, mammal bone; ▲, dentine: from a horizon between the Upper and the Middle tuff in the Kabuh formation at the Pucung excavation site. Correlation coefficient is calculated excepting the data on dentine.

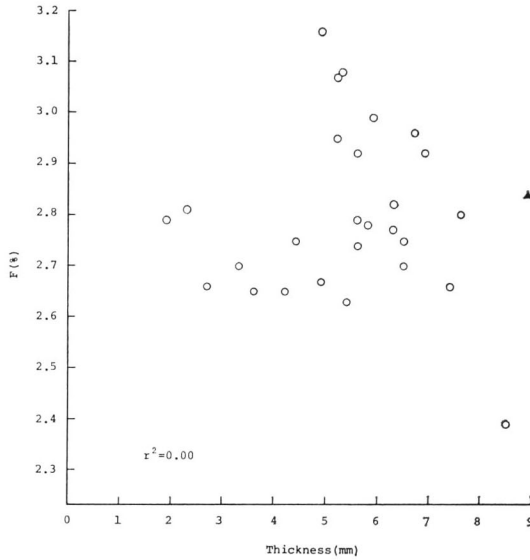


Fig. 9. Fluorine content of compact bone and dentine plotted against thickness of the specimen at the analysed part (exact figures given in Table 8). ○, vertebrate bone; ▲, dentine: from a horizon between the Middle and the Lower tuff in the Kabuh formation at the Brangkal excavation site. Correlation coefficient is calculated excepting the data on dentine.

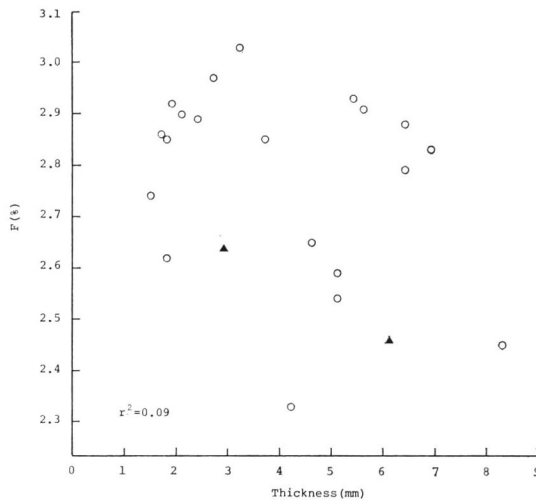


Fig. 10. Fluorine content of compact bone and dentine plotted against thickness of the specimen at the analysed part (exact figures given in Table 10). ○, vertebrate bone; ▲, dentine: from a horizon just below the Lower Tuff in the Kabuh formation at the Brangkal excavation site. Correlation coefficient is calculated excepting the data on dentine.

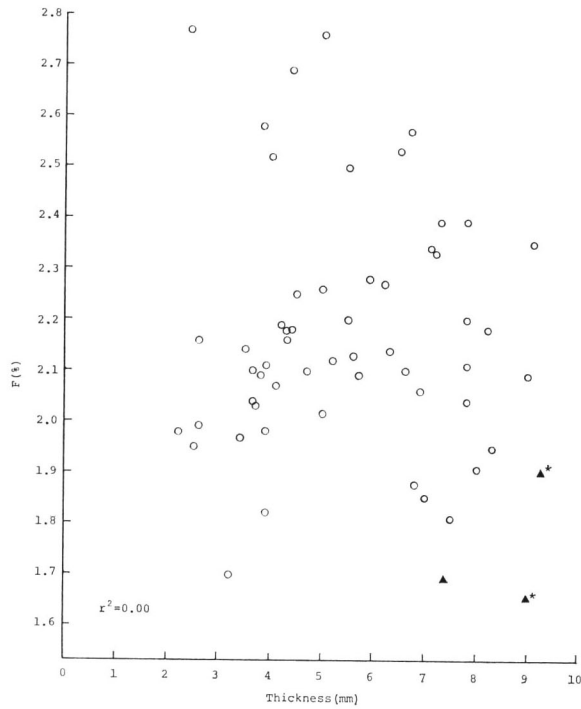


Fig. 11. Fluorine content of compact bone and dentine plotted against thickness of the specimen at the analysed part (exact figures given in Table 11). ○, vertebrate bone; ▲, dentine: from the Grenzbank zone at the Brangkal excavation site. Correlation coefficient is calculated excepting the data on dentine.

* Thickness data are the minimum estimates, because the inner portions of the two tusk specimens had come off.

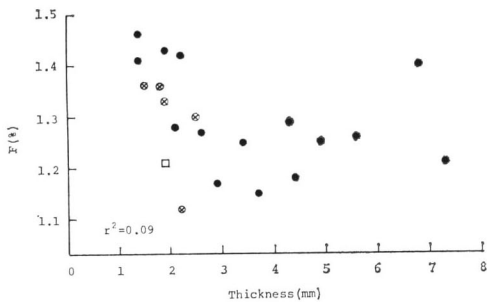


Fig. 12. Fluorine content of compact bone and horn core plotted against thickness of the specimen at the analysed part (exact figures given in Table 12). ⊗, tortoise shell; ●, mammal bone; □, horn core: from a horizon above Tuff 11 in the Pucangan formation at the Cengklik excavation site.

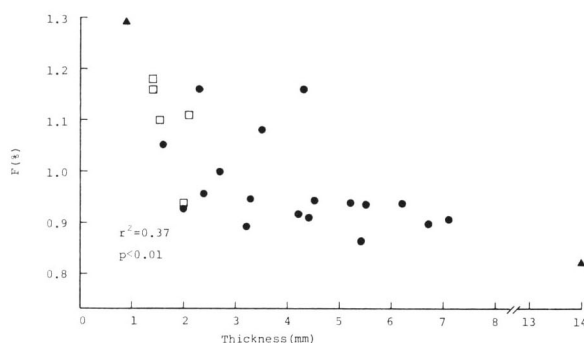


Fig. 16. Fluorine content of compact bone and antler and of dentine plotted against thickness of the specimen at the analysed part (exact figures given in Table 16). ●, mammal bone; □, antler; ▲, dentine: from a horizon between Tuff 4 and Tuff 3 in the Pucangan formation at the Pondok excavation site. Correlation coefficient is calculated excepting the data on dentine.

Table 17. Fluorine content of compact bone and dentine from the same fossil jaw. Data on *Cervus zwaani* from MATSU'URA (this study), and the other from PARKER *et al.* (1974).

Specimen	Age	F (%)	
		Bone	Dentine
<i>Cervus zwaani</i>	Pleistocene	1.11*	0.947**
<i>Diceratherium</i>	Miocene	3.31	3.24
<i>Subhyracodon</i>	Oligocene	2.95	2.67
Titanothere	Oligocene	2.37	2.63
<i>Coryphodon</i>	Eocene	2.15	2.32

* Thickness=1.7 mm.

** Thickness=6.2 mm.

more than one-fifteenth of the time interval that the vertebrate remains used here have experienced (see Figs. 6 & 21, Chapter VIII). Then, small changes in burial environments within a stratum would probably have a significant effect on the fluctuation in fluorine content of bones. For instance, the rock facies appears to be more uniform in the vertebrate beds of the Pucangan formation than in those of the Kabuh. In regard to compactness of deposit, Grenzbank which consists of sand and sandstone with granule and pebble conglomerates combined, may be the most fluctuating (see Fig. 11). The condition of the bones (i.e. intact or fragmentary) could also be a factor on their fluorine uptake, but it is a difficult question to answer when the fossil became fractured. Anyhow available information is still insubstantial for evaluating the contribution made by various parameters except thickness of bone, and I do not wish to engage in idle speculation at this time.

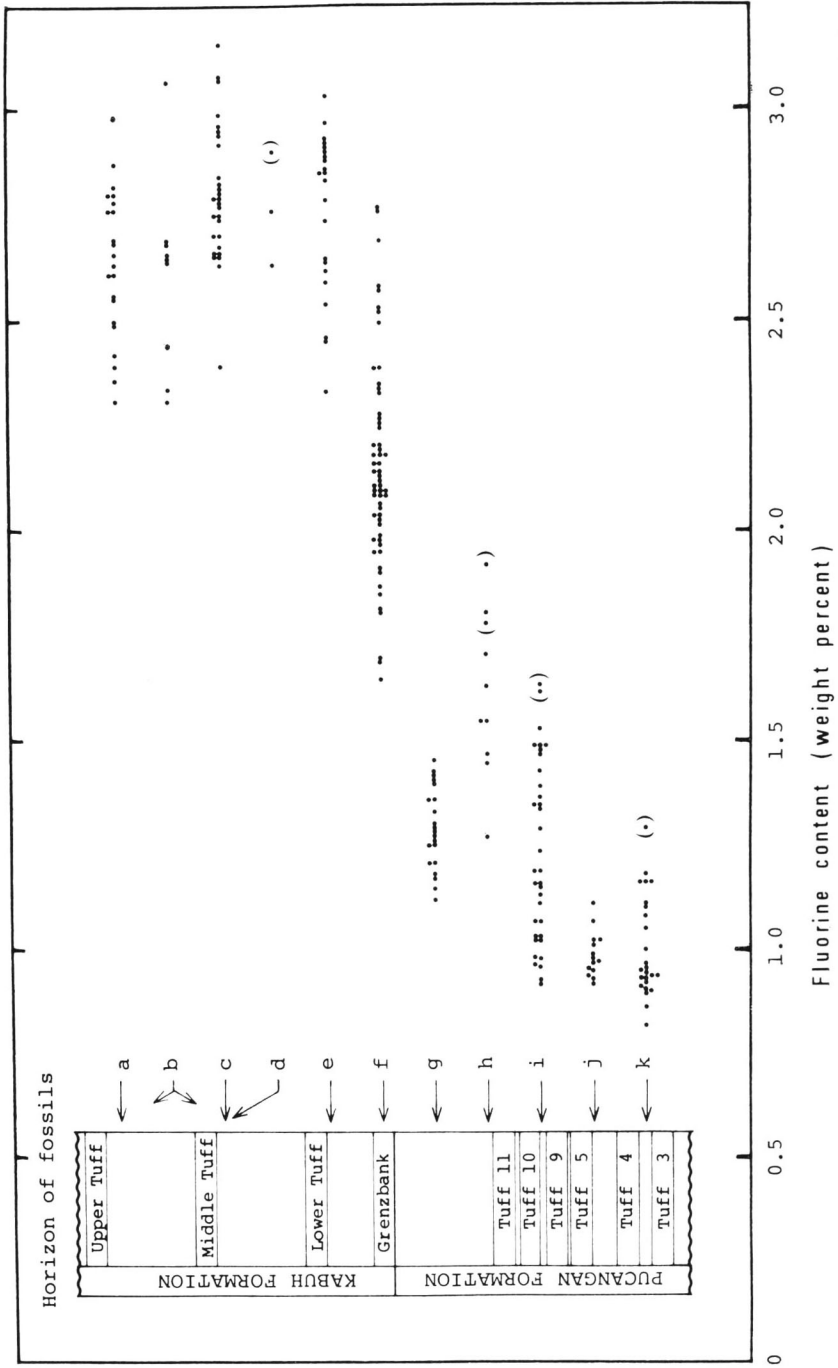


Fig. 17. Fluorine content in *Substantia compacta* of bone and antler, and in dentine from fossil vertebrate specimens unearthed *in situ* from various horizons in the Kabuh and Pucangan formations in the Sangiran area. Data for very thin (<1 mm thick) specimens are shown in parentheses. Exact figures given in Tables 5 to 16.

Table 18. Comparison of fluorine content of vertebrate fossils from Sangiran.
See Fig. 17 & Tables 5-16.

Horizon	F (%)		Pucangan Formation					Kabuh Formation			
	Range	Mean S.D.	T. 4 T. 3	just below T. 5	T. 10 T. 9	above T. 11 (J.)	above T. 11 (C.)	Grenz- bank zone	just below L.T.	M.T. L.T.	U.T. M.T.
Kabuh Formation	Upper T. Middle T.	2.31 2.98	2.64 0.18	>>>	>>>	>>>	>>>	>>>	>>>	v	∇
	Middle T. Lower T.	2.39 3.16	2.80 0.16	>>>	>>>	>>>	>>>	>>>	>>>	—	
	just below Lower T.	2.33 3.03	2.76 0.19	>>>	>>>	>>>	>>>	>>>	>>>		
	Grenzbank zone 2.77	1.65 2.77	2.15 0.25	>>>	>>>	>>>	>>>	>>>	>>>		
	above Tuff 11 (Cengklik) 1.46	1.12 1.46	1.29 0.10	>>>	>>>	—	∇ ∇				
	above Tuff 11 (Jagan) 1.92	1.27 1.92	1.62 0.19	>>>	>>>	>>>					
Pucangan Formation	Tuff 10 Tuff 9	0.921 1.64	1.23 0.21	>>>	>>>						
	just below Tuff 5	0.917 1.11	0.986 0.055	—							
	Tuff 4 Tuff 3	0.821 1.29	1.00 0.12								

— not significant
 > significant at 5% level
 >> significant at 1% level
 >>> significant at 0.1% level
 (by Mann-Whitney U-test)

Now, in Fig. 17 the fluorine measurements on the Sangiran specimens are collected and illustrated graphically according to their horizons. Statistical tests of the disparity in fluorine content between horizons yielded the results tabulated in Table 18. These tests were made without regard to the difference between the sample means of bone thicknesses; and the results should represent not the differences of “the fluorine content expected of bone samples with a given thickness”, but the differences in practice since the pilot series of fossil samples for each horizon was chosen with the intension of reflecting the dispersion in thickness.

The results shown in Fig. 17 and Table 18 are interesting and were what had not been foreseen. Any of the bone sets from the younger (upper) formation, Kabuh, has shown significantly higher fluorine content than all of those from the older Pucangan formation (Table 18). Admittedly the bulk of fluorine in fossil bone and tooth is due to diagenetic gains from the burial media by the apatite structure (see Chapter III). As Fig. 17 shows, however, the average concentration of this element in the fossils form Sangiran fails to vary monotonically with age. It appears to reach a maximum in the specimens from the lower part of the Kabuh excluding Grenzbank, and a minimum in those from the two layers between T3 and T5 in the Pucangan.

As far as I know, this instance is the first outstanding one contrary to the usual

trend that the average fluorine content of buried skeletal material from neighboring sites exhibits a steady increase with their antiquity. Considering the Pucangan samples are slightly thinner than the Kabuh samples, the disparity seen in Fig. 17 or Table 18 between the fluorine data for the two formations would not be reduced after adjustment of difference in thickness of sample. The outcome found for the Sangiran specimens is the unexpected, yet not surprising because the Kabuh formation is very different from the Pucangan in such important burial environments as facies of stratum, permeability of matrix (see Chapters I & IV), and amount of soil fluorides (see the section after next).

Although not uniquely related to age, the fluorine data presented here can be used as comparative series for checking up the observed or alleged association of a Sangiran hominid with a particular bed and in some cases for allowing inferences regarding the source horizons of human remains from the Sangiran area.

4. Phosphate Measurements.

Alternative interpretation of the phenomenon observed in Fig. 17 might be suggested that the more introduction of adventitious mineral matter (to act as a weight

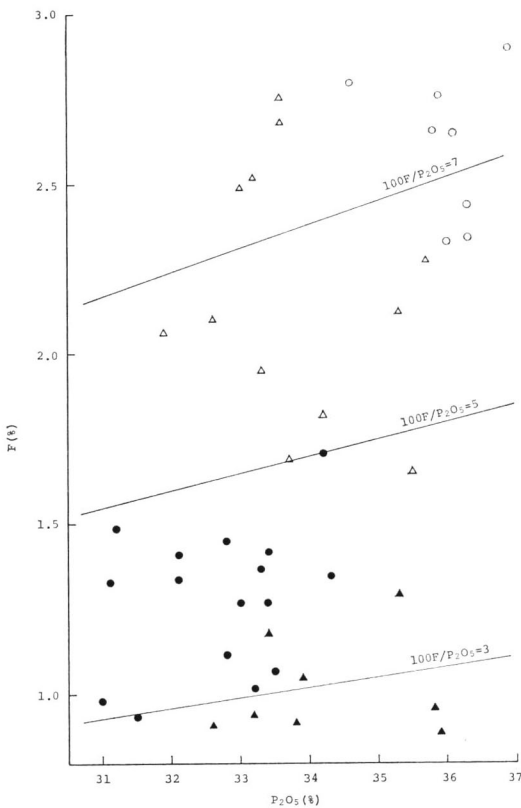


Fig. 18. Fluorine and phosphate analyses of compact bone and antler and of dentine from Sangiran. ○, from Kabuh excluding Grenzbank; △, from Grenzbank zone; ●, from Pucangan layers above Tuff 9; ▲, from Pucangan layers below Tuff 5.

in sample) into fossils from the Pucangan formation should induce the lower fluorine values for them. However, this can not be the case since all the samples involved do not appear to have suffered such kind of contamination. To confirm this, phosphate in bone was determined, because the concentration of this constituent provides a convenient measure of the apatite present in fossil bone.

Samples were selected to almost represent the variety of fluorine content exhibited in Fig. 17, and assayed for phosphate with the result shown in Fig. 18. The P_2O_5 measurements range from 31.9% to 36.9% in the Kabuh samples, and from 31.0% to 35.9% in the Pucangan samples; the difference between the two groups (Kabuh, $n=20$, $\bar{x}=34.8\%$; Pucangan, $n=24$, $\bar{x}=33.2\%$) is small and not significant. It is noticed that the phosphate analyses of fossils from the Grenzbank zone of the Kabuh and from the Pucangan layers above Tuff 9 are located in lower ranges of the analyses on the Kabuh and Pucangan samples respectively. Yet, these variations are too minor to account for Fig. 17. Phosphate content of the Sangiran specimens (Fig. 18) is of the same level as reported by BROPHY & NASH (1968), SEITZ & TAYLOR (1974), MATSUBARA (1980), and others for fossilized dentine and compact bones which are thought to have suffered little intrusion of foreign mineral matter.

The result presented above is to substantiate the conclusion that Fig. 17 reflects the degree of fluorination in the bone apatite structure. The difference in fluorination between fossil bones from Kabuh and Pucangan is manifested by Fig. 18 (note isoplethic lines of the ratios $100F/P_2O_5$).

5. Fluoride Content in Soil Extracts and the Rate of Fluorine Uptake by Buried Bone.

As mentioned earlier, the accumulation rate of fluorine in bone is governed by the rate at which OH^- ions in bone apatite are substituted by F^- ions, and by the rate at which fluoride ions diffuse into the bone.

Now suppose the thickness of a fossil bone approaches to zero, then diffusion is not a factor. Let the surrounding fluoride concentration be held constant; since the conversion of hydroxyapatite to fluorapatite is practically irreversible (see HAGEN, 1973; TSE *et al.*, 1973), the rate of increase in fluorine content ($F\%$) of bone apatite structure is thus to be proportional to the amount of apatites that remain to be fluorinated, and may be given by:

$$\frac{dF}{dt} = k \times (3.77 - F)$$

where k is the rate constant of the reaction that fluorine becomes fixed in the apatite crystals, and t , the elapsed time. Upon integrating the above equation and setting $F=0$ at $t=0$, we have

$$\ln \left(\frac{3.77 - F}{3.77} \right) = -kt \quad (i)$$

Fig. 19 represents the relations between k values and the amounts of matrix-soil

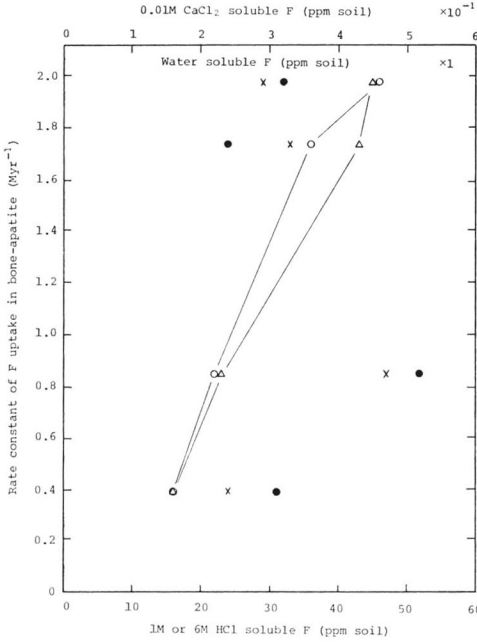


Fig. 19. Soil fluoride concentrations and the rate constant of fluorine uptake in bone-apatite. For calculation of the rate, see text. ×, 0.01 M CaCl₂ sol.; ●, water sol.; ○, 1M HCl sol.; △, 6M HCl sol.

Table 19. Fluoride concentrations of matrix of fossil vertebrate bed. Samples were air-dried and sieved to <0.5 mm.

Locality	Stratigraphic provenance*	Lithological note	F (ppm soil) soluble in			
			0.01 M CaCl ₂ [†]	Water ^{††}	1 M HCl ^{††}	6 M HCl ^{††}
Pucung excavation site	U.T.-M.T. Kabuh	sand & gravel with pumice ball	0.29	3.2	46	45
Brangkal excavation site	M.T.-L.T. Kabuh	sand with granule & pebble	0.33	2.4	36	43
Brangkal excavation site	Grenzbank zone	sand & sandstone with gravel, and conglomerate	0.47	5.2	22	23
Bukuran excavation site	T. 10-T.9 Pucangan	silt & clay	0.24	3.1	16	16

* See Fig. 6.

† 1:3 solid-weight/solution-volume, 3-day diffusion period at room temperature (see LARSEN & WIDDOWSON, 1971).

†† 1:20 w/v, 5-day contact time at room temperature.

fluoride extracted with four types of solvents (Table 19) for some of the fossil bearing horizons in Sangiran. The *k* values were approximated by substituting *t* and *F* in eq. (i) with the age found from Fig. 21 (see Chapter VIII) and the fluorine content value at thickness=0 extrapolated from the regression equation of fluorine content

on thickness of compact bone (see Figs. 8–16) for each horizon involved, respectively.

Soil fluoride extracted with water or saline solutions has been considered to be the source of the element to plants (YAMADA & HATTORI, 1979; YUIDA, 1980, *in lit*). Their correlations to k , however, are poor as seen in Fig. 19. YUIDA (*ibid.*) has suggested that soil fluoride which will be solubilized over a period of geological time may be available for buried bones. It is supported by the present experiment, where the concentration of 6 M HCl-soluble fluoride is found to be best correlated with k ($r=0.99$, $0.01 < P < 0.02$).

B. Substantia Spongiosa

Table 20 shows that the fluorine content measured on a cancellous tissue sample which is contaminated from extraneous mineral grains or soil particles may be corrected on the basis of phosphate measurements so as to compare roughly with the fluorine content of the compact tissue of the same bone, at least as far as vertebrate fossils from Sangiran are concerned.

OAKLEY (1963, p. 113) has stated that fluorine “is adsorbed at slightly higher rates in spongy bone or antler” than in compact bone or antler. Table 20, however, fails to support this statement.

Table 20. Fluorine and phosphate contents of compact bone and of spongy bone with varying amounts of infiltrated mineral matter.

Specimen	Formation	compacta			spongiosa			Ratio of F cor., sp./ compacta	
		T* (mm)	F (%)	P ₂ O ₅ (%)	F (%)	P ₂ O ₅ (%)	F (%) cor.**		
mammal	Kabuh	1.8	2.80	34.6	1.46	18.8	2.69	0.96	
mammal	Kabuh	7.1	2.76	35.9	1.91	26.9	2.55	0.92	
mammal	rib	Kabuh	2.2	2.66	35.8	1.92	25.4	2.71	1.02
cervid	metapodial	Pucangan	2.3	1.45	32.8	1.42	30.9	1.51	1.04
mammal	rib	Pucangan	2.2	1.42	33.4	1.25	29.6	1.41	0.99
mammal	vertebra	Pucangan	1.4	1.41	32.1	1.10	27.7	1.27	0.90
tortoise	shell	Pucangan	2.5	1.33	31.1	1.00	21.0	1.48	1.11
mammal	rib	Pucangan	2.6	1.27	33.0	0.515	13.0	1.31	1.03
tortoise	shell	Pucangan	2.2	1.12	32.8	0.639	21.6	0.970	0.87
mammal	rib	Pucangan	2.5	0.982	31.0	0.352	11.9	0.917	0.93
<i>C. hippelaphus</i>	antler	Pucangan	3.2	0.893	35.9	0.455	18.5	0.883	0.99
							Mean	0.98	
							S.D.	0.07	

* Thickness of compact tissue.

** Corrected to P₂O₅ content of compact tissue from the same specimen.

C. Enamelum

ROWELS (1967) and PARKER *et al.* (1974) have demonstrated that the amount of fluorine is lower in enamel than in dentine from the same specimen of recent teeth. The proportion of fluorine content in dentine (F: 0.008–0.16%) to enamel in each of

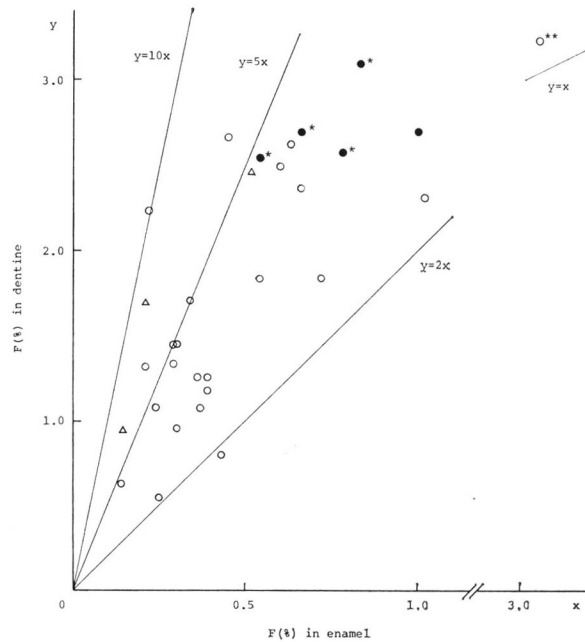


Fig. 20. Fluorine content of dentine and enamel of fossil teeth with little contamination of adventitious mineral matter. References: open circle, PARKER *et al.* (1974); solid circle, Tables V & VII in WEINER *et al.* (1955); open triangle, MATSU'URA (this study).

* 'F (%) in dentine' is for dentine+cementum sample.

** Exception to the low fluorine content in enamel.

these recent teeth ranges 1.1 to 5.9. The proportions in fossil teeth, as Fig. 20 shows, vary approximately from 2 to 10 with the exception of one data marked with double asterisk. As a result of large differences in physical properties, diagenetic gains in fluorine are much less in enamel than in dentine (see Chapetr III, B). This may explain the finding that the aforementioned proportions are generally larger in fossil teeth than in recent specimens.

Most of fluorine in enamel, fossil or recent, occurs in the surface layers (GLOVER & PHILLIPS, 1965; BRUDEVOLD & SÖREMARK, 1967). Therefore thicker enamel is expected to carry a lower average amount of fluorine. This tendency can be observed for Sangiran material in the following way:

ratio of thickness enamel/dentine	ratio of fluorine enamel/dentine
0.15	0.21
0.18	0.15
0.28	0.12

(No measurements for thickness have been reported with the other data in Fig. 20)

The thickness dependent variation in fluorine content of enamel is most likely to be violent. Consequently we must severely consider the thickness factor when fluorine-dating a fossil hominid using enamel samples. Moreover, it can hardly be imagined that the enamel part of a precious human specimen is submitted to the relative dating processes together with animal samples suitable for comparison (teeth provide most important information on taxonomy in mammalian palaeontology). Hence at this stage, enamel may not be the object of the fluorine dating method.

VII Consideration of the Stratigraphic Positions of Hominid Remains from Sangiran Based on Fluorine Analyses

Table 21 gives the results of analysing compact bone samples of *Pithecanthropus* VII & VIII, and published fluorine analyses for Sangiran hominid remains. These analyses are compared with the standard data presented in Chapter VI.

The fluorine content values of Sangiran 1, 2, 4 and 5 (Table 21) may have been diluted to a considerable extent by infiltrated minerals or other foreign matter, judging from the accompanying P_2O_5 measurements. They should be treated after being compensated for the contamination. Those four human fossils are discussed in the second

Table 21. Fluorine and phosphate contents in fossil hominid remains from Sangiran. Data for Sangiran 1-6 are from OAKLEY *et al.*, (1975)*, and for Sangiran 12, 17 from MATSU'URA (this study).

Specimen	F (%)	P_2O_5 (%)
Brit. Mus. Cat. No.		
Sangiran 1** Pithecanthropus B	0.61	9.7††
Sangiran 2 Pithecanthropus II	1.15	22.5††
Sangiran 3 Pithecanthropus III	2.93	38.1††
Sangiran 4 Pithecanthropus IV	1.33	20.8††
Sangiran 5 Holotype of <i>P. dubius</i>	0.56	10.6††
Sangiran 6† Holotype of <i>M. palaeojavanicus</i>	2.32	35.2††
Sangiran 12 Pithecanthropus VII	2.74	35.3
Sangiran 17 Pithecanthropus VIII sample 1‡	2.73	34.6
sample 2‡	2.61	36.7

* British Museum's Catalogue of Fossil Hominids (Part III) lists analytical data on Sangiran 8, i.e., *Meganthropus* B (see OAKLEY *et al.*, 1975, p. 111); but this entry is incorrect and should be deleted (MOLLESON, *in lit* 1982).

** Sangiran 1 consists of S 1a (maxilla) and S 1b (Pithecanthropus B). The analytical data on Sangiran 1 is for Pithecanthropus B (MOLLESON, *in lit* 1982).

† The analytical data on Sangiran 6 is for the right corpus of the mandible found in 1941 (MOLLESON, *in lit* 1982), not for the left gonial portion that is also included in Sangiran 6 (see OAKLEY *et al.*, 1975).

†† Calculated from F content and 100F/ P_2P_5 ratio.

‡ From two different chips of bone.

division (B in this chapter). The first division deals with Sangiran 3, 6, 12 and 17, the analysed samples of which are free from the contamination problem.

A. *Pithecanthropus* III, VII, VIII; *Meganthropus*, Holotype

1. *Pithecanthropus* III.

Pithecanthropus Skull III, a juvenile calvaria, was picked up in a case containing fossils collected from the Trinil (Kabuh) beds of Sangiran (VON KOENIGSWALD & WEIDENREICH, 1938; WEIDENREICH, 1945, p. 13–14). The fluorine content of Skull III, on referring to Fig. 17, suggests that this fossil hominid came from the Kabuh formation, and plausibly from a horizon lying below the Middle Tuff and above Grenzbank. This is compatible with views on its stratigraphic position given by SARTONO (1975) and by Indonesia-Japan Research Cooperation Programme—CTA-41 (1979); that is, a lower level in the Kabuh formation, and a horizon between the Middle and the Lower Tuff in the Kabuh, respectively.

2. *Pithecanthropus* VII and VIII.

The deposits that were reported to have contained *Pithecanthropus* VII (SARTONO, 1967) and VIII (SARTONO, 1972) skulls have both been assigned by ITIHARA *et al.* (Indonesia-Japan Research Cooperation Programme—CTA-41, 1979) to a horizon between the Middle and the Lower Tuff of the Kabuh formation. The fluorine test on the two hominid material has not raised any objection against this assignment.

3. *Meganthropus palaeojavanicus*, holotype.

A massive human mandible, which represents the holotype of *Meganthropus palaeojavanicus*, was sent to VON KOENIGSWALD from Sangiran in 1941. He presumed afterwards that this specimen had been found in the lower horizon, i.e., part of the Pucangan formation (VON KOENIGSWALD, 1947, 1950, 1968a). The fluorine data, however, fails to support this presumption, and leads me instead to maintain that the 1941 mandible is probably from Grenzbank, the lowermost layer of the Kabuh formation. From this layer, another mandible of *Meganthropus* (*Meganthropus* B) was found in 1952 as communicated by Marks (1953).

VON KOENIGSWALD (1968b) reports that a fragment belonging to *Meganthropus* has 2.48% F. Although we are not well informed with which specimen this assay is concerned, I read from VON KOENIGSWALD (1968b, 1973, 1975) that the specimen would come from the Sangiran area and that it should not be the 1941 mandible. Making reference to Fig. 17, we find it difficult to avoid the conclusion that this fragment of *Meganthropus* also has its origin in the Kabuh formation.

B. *Pithecanthropus* B, II, IV; *P. dubius*

Upon correcting the fluorine measurements for *Pithecanthropus* B, II, IV and *P. dubius* (S5) to 33.9% P₂O₅ (the mean of phosphate analyses of compact bone samples from Sangiran; see Chapter VI, A-4), we can read their fluorine content as follows:

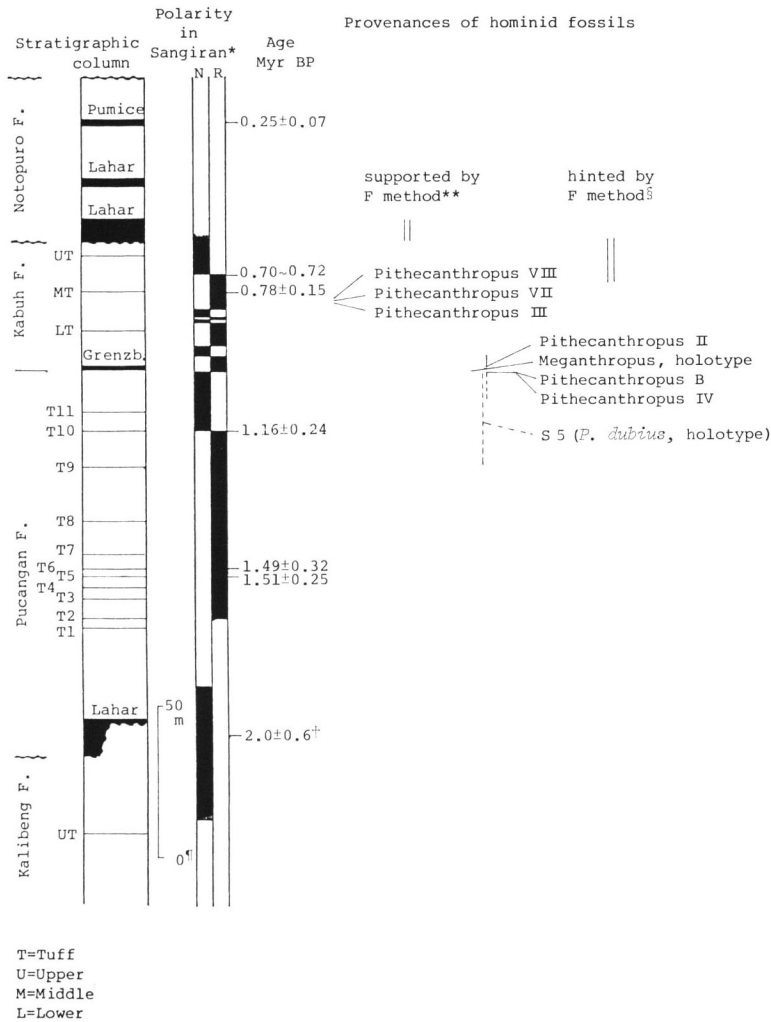


Fig. 21. Correlation of fossil hominid horizons with ages and polarities observed in the formations exposed in the Sangiran area.

* After SHIMIZU *et al.* (1983).

** Positions inferred by geological investigations and supported by the fluorine method.

§ Positions suggested by the fluorine method with geological and other data tempered (see text).

† K-Ar age (NISHIMURA *et al.*, 1981). All other ages (with $\pm 2\sigma$) were determined by the fission track method (SUZUKI *et al.*, 1983).

‡ Lahar, pumice and Grenzbank layers are not to scale.

- Pithecanthropus B, 2.1₃% F (1.7₉-2.4₇);
 Pithecanthropus II, 1.7₃% F (1.4₆-2.0₁);
 Pithecanthropus IV, 2.1₇% F (1.8₂-2.5₁);
P. dubius (S5), 1.7₉% F (1.5₀-2.0₈).

The range shown in parentheses is $\pm 16\%$ of the corrected F value for each specimen, giving a possible fluctuation ($\pm 2\sigma$) which is derived both from the variation seen in Table 20 and from that in P₂O₅ content of compact tissues of Sangiran specimens (see Fig.18).

1. Pithecanthropus II.

The second skull of *Pithecanthropus* came in 1937 from near Bapang (Fig. 5) on the left bank of the river Cemoro (VON KOENIGSWALD, 1938, 1940). This surface find was estimated to be washed out of the sandstones and conglomerates, a lower part of the Kabuh formation (e.g. VON KOENIGSWALD 1947; DE TERRA, 1943). ITIHARA *et al.* (IJRCP—CTA-41, 1979) have referred its original stratigraphic position to the Kabuh layers between the Middle and the Lower Tuff. The fluorine content, however, implies that the specimen was derived from Grenzbank. Reassay on Pithecanthropus II using a compact bone sample would settle the above discrepancy.

2. Pithecanthropus B and IV; *P. dubius*, holotype.

These hominid fossils were found by native collectors from 1936 to 1939 (VON KOENIGSWALD, 1937; WEIDENREICH, 1945; HOOIJER, 1951). Although their source horizons could not be definitely deduced, VON KOENIGSWALD (1950, 1968b, and others) claimed that they should have their origin in the Pucangan layers. I deduce from comparisons of the fluorine data that Pithecanthropus B and IV came probably from Grenzbank or possibly from the uppermost part of the Pucangan, and *P. dubius* from Grenzbank or upper Pucangan beds.

VON KOENIGSWALD (1968b) reports that the mandible of *modjokertensis* [i.e. Pithecanthropus B] has 2.13% F. This value happens to coincide exactly with the estimate in this work. If that measurement (2.13%) is one obtained using a compact bone sample with little or no contamination, the possibility of Pithecanthropus B being from Grenzbank is stronger.

Conclusions on the stratigraphic provenances of the Sangiran remains listed in Table 21 are summarized in Fig. 21.

Recently HOLLOWAY (1981) has revised the Indonesian *H. erectus* brain endocasts with the following result:

- Pithecanthropus VII=1,059 ml;
 Pithecanthropus VIII=1,004 ml;
 Pithecanthropus II=813 ml;
 Pithecanthropus IV=908 ml.

This is compatible with the chronological sequence of the hominid fossils shown in Fig. 21.

VIII Ages for the Sangiran Hominids

SUZUKI *et al.* (1983) treated 21 samples of (pumice) tuff layers and lahar, and 2 Javan tektites collected *in situ* from the late Cenozoic Group exposed in Sangiran; and have reported fission track dates for the 'primary' zircon grains obtained from the Pumice Tuff (Notopuro), the Middle Tuff (Kabuh), Tuff 10, Tuff 6, Tuff 5 (Pucangan) and the Lower Tuff 1 (Kalibeng), and fission track dates for the 2 tektite specimens (see Fig. 6). These dates (with $\pm 2\sigma$) excluding the one for the Lower Tuff 1 are shown in Fig. 21, which also records the results of a palaeomagnetic study by SHIMIZU *et al.* (1983), the most extensive one carried out in the Sangiran area, I suppose, as yet.

Here I must refer to the results published by NISHIMURA *et al.* (1980) of fission track dating of the tuffaceous units within the Kabuh and Pucangan formations of Sangiran. They used for the age calculation the youngest peak of the scattered fission track ages of zircon crystals separated from each unit; and have given $0.47 \text{ Myr} \pm 8.5\%$ (1σ) and $0.51 \text{ Myr} \pm 9.6\%$ to two tuff layers in the upper half of the Kabuh, and $0.56 \text{ Myr} \pm 8.8\%$ and $0.67 \text{ Myr} \pm 9.6\%$ to two tuffs in the upper Pucangan. On comparing these age values of NISHIMURA *et al.* (although the horizons of the samples of NISHIMURA *et al.* for dating have not been well correlated with Fig. 6) with those reported by SUZUKI *et al.* (1983), we may be impressed by the magnitude of the present dilemma where one fission track dating group produces a series of dates which is about two thirds of that produced by the other group. Both have adopted $7.03 \times 10^{-17} \text{ yr}^{-1}$ as a fission decay constant of U-238, and agreed as to the dates for tektites from Sangiran (0.67 Myr by NISHIMURA *et al.*, 1980; $0.70\text{--}0.72 \text{ Myr}$ by SUZUKI *et al.*, 1983). Accordingly the present discrepancy must be due to the experimental approach for dating zircons.

As GLEADOW (1980) has pointed out, fission track dating methods for zircons of very young geological age encounter particular difficulties such as follows:

- 1) The very low track etching rate and the low density of spontaneous tracks make it difficult to achieve an optimal track etching.
- 2) The anisotrophical effect of a crystal on the revelation of tracks is high.

SUZUKI *et al.* (1983) made due consideration and checking of these factors, whereas NISHIMURA *et al.* (1980) did not mention to what extent these factors had been checked. Still, there is another matter to be noted, to which I attach most importance for the rating of their dating results; that is the density of the induced fission tracks with the thermal neutron bombardment. In the study by SUZUKI *et al.*, this density was less than $5 \times 10^5/\text{cm}^2$, i.e., less than 50 tracks in a 100μ square field (the density of fossil tracks was about a tenth), a level of density reasonable for the track counting. On the other hand the induced track density in the study by NISHIMURA *et al.* was $6\text{--}9 \times 10^7/\text{cm}^2$, i.e., 6,000–9,000 tracks per 100μ square. Such extraordinarily high density involves the presence of TINT (Track-in-track) tracks, assuming the length of fission track is around 10μ . Provided that the TINT phenomenon resulted in an apparent gain in the induced track density, then a misleading underestimate of the age would

have been generated. Anyhow, the extra high track density might make it impossible to distinguish without confusion the exact number of tracks.

For the reasons mentioned above and below, I have adopted in this study the age estimates given by SUZUKI *et al.* (1983).

a) Javites from Sangiran belong to a widespread shower of tektites which affected Indo-China and an Oceanic strewn field. Many age determinations by K-Ar or fission track method on these tektites group around 0.7 Myr (ISAAC, 1972; FLEISCHER *et al.*, 1975). The stratigraphic position of javites in the Sangiran area has been confirmed by the CTA-41's fieldwork to be between the Upper and the Middle Tuff of the Kabuh formation (see Fig. 6). This position is to be at the boundary between the Brunhes normal and the Matuyama reversed epoch (Fig. 22). The palaeomagnetic data for Sangiran (Fig. 21) are not contradictory to the above.

b) According to KADAR (1979, verbal communication), the boundary of the Kalibeng and Pucangan formations lies around the boundary between N21 and N22 of the planktonic foraminiferal zones. NINKOVICH & BURCLE (1978) estimated the base of the Pucangan beds to be at 1.9–2.1 Myr through the diatom analysis. Considering a published K-Ar date (marked with † in Fig. 21) and the aforementioned all at once, the normal polarity observed around the lahar layer in the basal Pucangan may be assigned to either the Olduvai or the Réunion event (see Fig. 22). The fission track ages reported in SUZUKI *et al.* (1983) are consistent with this view.

c) Short normal layers are observed continually in the lower or the lowermost part of the Kabuh formation and the uppermost part of the Pucangan (Fig. 21). We can find only the Jaramillo event in this period of the standard geomagnetic polarity time scale; yet the time scale is being modified and addition of some events is suggested (SHIMIZU *et al.*, 1983). In any case, with two key dates of 0.7 Myr (Brunhes/Matuyama boundary) for the upper-middle Kabuh and c. 2 Myr for the basal Pucangan, the age of the Kabuh/Pucangan boundary is deducible from the thickness of the strata to be c. 1 Myr. The sedimentation rate involved is about 100 m/million years. This is supported by a fission track date of $2.99 \pm 0.47(2\sigma)$ Myr which SUZUKI *et al.* (1983) report for the Lower Tuff 1 (Kalibeng) lying about 100 m below from the top of the Kalibeng formation.

d) Tuff samples collected from the Kabuh formation near the sites of Pithecanthropus VI and Pithecanthropus VII have yielded four K-Ar dates of 0.781–0.908 Myr (average, 0.83 Myr) (JACOB, 1973, 1978). These dates are compatible with those quoted in Fig. 21.

In view of these discussions it is concluded that the source horizons of the Sangiran hominid remains treated in this study may span the time from c. 0.8 to c. 1.1 or possibly to 1.3 Myr, all belonging to the Lower Pleistocene.

Some dates which were determined on samples obtained from several sites in Java other than Sangiran have been applied by correlation to the sediments in Sangiran. Among these dates (excluding ones for tektites) we find those described below.

a) The site of *Pithecanthropus (Homo) modjokertensis* at Perning was reported to be associated with the Jetis fauna (VON KOENIGSWALD, 1936, 1940). A pumice tuff sample collected a few metres below the finding spot has given a K-Ar age of 1.9 ± 0.4 Myr (JACOB, 1972). This estimate has been applied to the Sangiran Pucangan layers (VON KOENIGSWALD, 1975; SARTONO, 1975; JACOB, 1975b; CRONIN *et al.*, 1981; and others) on the ground that these layers were said to contain the Jetis fauna and that *Pithecanthropus B* and *Pithecanthropus IV* which were believed to be derived from the Sangiran Pucangan formation were described as belonging to the same species as represented by the type specimen of *modjokertensis* from Perning. As to fauna, VAN HEEKEREN (1972) also states that the Pucangan formation at either Perning or Sangiran yields guide fossils of the Jetis fauna. Recent evidence (AIMI & AZIZ, 1983), however, casts a doubt on the validity of the Javanese vertebrate stratigraphy that has ever been used (see Chapter I). And as of now it is impossible to correlate geologically Sangiran with Perning (KUMAI, 1981, personal communication). Taking account of the aforementioned and the large error (0.4 Myr) in the age determination, we can not adopt 1.9 Myr as an age estimate for the Sangiran Pucangan beds.

Lately SARTONO *et al.* (1981) have given findings on fossil vertebrate fauna in the *modjokertensis* site different from those described by VON KOENIGSWALD (1940), and also questioned the assignment of 1.9 Myr to the holotype of *modjokertensis* itself.

b) Two leucitic-basalt samples from the Muriah volcanoes about 80 km north of Trinil have been dated at $0.495^{+0.10}_{-0.06}$ Myr (LIPPOLT in VON KOENIGSWALD, 1962) and 0.500 Myr (EVERNDEN & CURTIS, 1965) by the potassium-argon method. The basalt is said to be related with a "typische Trinilfauna" (VON KOENIGSWALD, 1962); yet no detailed reports of the faunal or stratigraphical correlations involved are available. The significance of the two dates for the Sangiran area remains uncertain.

IX Chronological Position of Sangiran Hominids in Human Evolution

This chapter outlines the chronological positions of some of the hominid fossils from the Sangiran area as compared with those from other regions, and gives some remarks on their palaeoanthropological implications.

A. Indonesia

The layers that are thought (or reported) to have yielded the Trinil remains and the Sambungmacan remains (see Chapter II) have been assigned to the lower half of the Kabuh formation (IJRCP—CTA-41, 1979; KUMAI, 1981, personal communication). Detailed correlations with the deposits of Sangiran would be unfeasible since the sedimentation rate in Trinil or Sambungmacan is much smaller than that in Sangiran. Perning (Mojokerto) site is difficult to make a good geological correlation with Sangiran site because of the differences in rock facies, and thus needs absolute dates measured on the primary minerals from tuffs (see Chapter VIII).

C. Europe

Age estimates have been obtained for fossils from e.g. Swanscombe, Arago, Petralona, Vertésszöllös and Bilzingsleben (CHERDYNTSEV *et al.*, 1965; SZABO & COLLINS, 1975; IKEYA 1980; HARMON *et al.*, 1980; HENNIG *et al.*, 1981; etc.). Any of the estimates that exceed 0.5 Myr is still controversial.

D. East Africa

Fig. 22 summarizes correlations between Sangiran and some hominid sites in East Africa. TOBIAS & VON KOENIGSWALD (1964) concluded that *Pithecanthropus* B and *Pithecanthropus* IV had strong resemblances to OH13 (Bed II '*Homo habilis*'), and the 1941 specimen of *Meganthropus* to OH7 (Bed I '*H. habilis*'); and suggested that the two pairs seem to be of the same grade of hominization respectively. Nevertheless their proposition is contrary to the chronology of these hominid material (Fig. 22). Another criticism is from LOVEJOY (1970) that TOBIAS & VON KOENIGSWALD (1964) are based much on some similarities of traits which are not relevant to taxonomy.

The Sangiran remains involved in Chapter VII (see Fig. 21) are comparable in age to the material from the upper Bed II to the lower Bed IV of Olduvai.

E. Note on *Meganthropus*

Among the Sangiran hominid fossils *Meganthropus* specimens have been the subject of much controversy and conjecture regarding their taxonomic status and place in human evolution (e.g. WEIDENREICH, 1945; ROBINSON, 1953, 1954; SARTONO, 1961; TOBIAS & VON KOENIGSWALD, 1964; LOVEJOY, 1970; VON KOENIGSWALD, 1973; CLARK, 1978). The variety of opinion is to be caused by lack of reliable age estimates and lack of available material (limited to jaws and isolated teeth). Besides no detailed description has been presented for some specimens which were claimed to be ascribed to *Meganthropus*. One of them is regarded by JACOB (see 1973) as a *Pongo* molar (see Table 1).

In 1978 the first cranium that is identified as *Meganthropus* though tentatively was discovered from the central part of the Sangiran dome (JACOB, 1980). The horizon which is said to have yielded the skull is assigned to the upper part of the Pucangan formation (Indonesia–Japan Joint Research Team, 1979). However, since the mid-dome area shows a complicated geological structure (see Chapter I), further investigations using e.g. the fluorine method would be needed to elucidate the above assignment. Phylogenetic studies of the skull want to be conducted with due consideration of chronological backgrounds.

Concluding Remarks

This study has clarified the stratigraphic provenances of some of the Sangiran human remains, and their relative time relations. Absolute ages of them have been inferred through the rating of the reported dating results of the sediments in the Sangiran area to be around 0.8 Myr for *Pithecanthropus* III, VII, VIII, and to be from c. 1.0

to 1.1 Myr for *Pithecanthropus* II, IV, B and the holotype of *Meganthropus palaeojavanicus*, and to be from c. 1.0 to c. 1.3 Myr for the holotype of *P. dubius*. This period of time suggested for these hominids is found to be not so long as hitherto guessed.

This work has shown that the fluorine dating method is promising for the fossil bones from the Sangiran area. Continuing investigations, analysing controlled samples of Sangiran hominid finds of unknown horizon, would be necessary components of informative studies in the chronology of human evolution. (Any proposal for my testing on human remains or for cross-checking analytical results would be highly appreciated.)

Synopsis

The Sangiran area in Central Java has yielded series of remains of early man that span over hundreds of thousand years. These hominid fossils were found, however, by chance by the local inhabitants, and their source horizons are difficult to locate. To solve the allocation problems the fluorine dating method is applied using as a pilot series a systematic collection of about 250 specimens of vertebrate fossils *in situ* from various horizons in this area.

Comparative data for the fluorine dating are presented by analysing compact bone and dentine samples. The data fail to follow the general trend that the average fluorine content of buried bones from the same region increases with age. Fossil bones from the younger formation, Kabuh, are found to have more fluorine than those from the older, Pucangan formation. Phosphate measurements prove that all the bone samples concerned underwent little intrusion of foreign mineral matter, and contamination problem should not involve the paradoxical result. This paradox is ascribed to the large difference in the burial environments between the two formations. Particular experiments are made on some of the burial matrices to examine the relations between the rate of fluorine uptake in bone and the fluoride concentrations in soil extracts, and give a hint to the state of fluoride available for buried bones.

Although not varying monotonically with age, the fluorine data presented can be used to deduce the possible source horizons of the Sangiran hominid remains, since they show highly significant differences between horizons or between parts of formation.

Variation in fluorine content of compact tissue with depth from the exposed surface of bone and the intra-specimen variation in fluorine content of cross segments of compact bone, are rated. The former is taken into consideration when analyses of hominid fossils are compared with the standard data for the fluorine dating; while the latter, when the standard data are constructed.

A series of tests on fossil bones from Sangiran shows that a correction procedure on the basis of phosphate measurements permits rough comparison of the fluorine content between cancellous regions contaminated by adventitious mineral or soil particles and dense cortical regions.

It is concluded that enamel is best excluded from the subject of the fluorine dating method.

As regards Pithecanthropus VII and VIII samples of which are analysed in this work and other Sangiran hominids on which chemical analyses including fluorine content have been published, their source horizons are suggested (or the reported associations of hominid specimens with particular beds are checked) by the fluorine dating approach as follows:

Pithecanthropus VIII	}	the lower part (between the Middle and the Lower Tuff) of the Kabuh formation
Pithecanthropus VII		
Pithecanthropus III		
Pithecanthropus II	}	Grenzbank (the basal layer of the Kabuh)
<i>Meganthropus</i> , holotype		
Pithecanthropus IV	}	Grenzbank to the upper part of the Pucangan formation
Pithecanthropus B		
<i>P. dubius</i> , holotype		

Through appraisalment of reported absolute dating results of sediments in the Sangiran area, these fossil hominids are inferred to span the age from c. 0.8 to c. 1.1 or possibly to c. 1.3 Myr, all assignable to the Lower Pleistocene. Here also outlined are their chronological positions in connection with early hominids from other regions in the world, especially from East Africa.

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