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CONTINENTAL PHOSPHATE DEPOSITS AND THE SEARCH FOR PHOSPHATES IN AUSTRALIA

by

O.N. WARIN

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INTRODUCTION

Since the discovery at the beginning of the century of the rich island phosphate deposits of Nauru and Ocean Islands in the Pacific Ocean and of Christmas Island in the Indian Ocean, most of Australia's increasing phosphate requirements have been met from these sources. The life of these deposits is severely limited however and the hope of finding other island deposits of equivalent size and richness is now very slender. Before the island deposits were discovered small deposits of phosphate rock were worked at a number of places on the continent. Production ceased at most of these centres in the early 1900s and there has been no impetus for further exploitation or for further prospecting for this type of deposit.

It is interesting to compare Australia's phosphate reserves with those of the United States of America. Australia uses at present about 1½ million tons of phosphate rock annually. We depend entirely on these external sources whose life, even in the unlikely event of a steady rather than a sharply rising demand for phosphate, will not be more than 30 years. The United States with an annual consumption of over 12 million tons, on the other hand, has estimated reserves of phosphate rock on the continent of over 40,000 million long tons* (Anon 1948). Most of this enormous tonnage is contained in what may be regarded geologically as two deposits; the residual deposits of Florida and the deposits of the Phosphoria Formation of the western states - Utah, Idaho, Wyoming and Montana. Itis also worthy of note that the aggregate thickness of the phosphorite beds in the latter formation is only 28 feet and that the thickest of them is nowhere more than 7 feet thick.

It is clearly important that deposits of this kind on the Australian mainland should not go undetected. What follows is a brief review of the different types of continental phosphate deposits; particular examples are described and their associated rocks noted as an aid to the recognition of similar deposits which may exist in Australia.

PHOSPHATES AND SUPERPHOSPHATES

History

Phosphatic materials have been used as fertilizers for a very long time; bird dung was used by the Carthaginians before 200 B.C. and its use was recommended by Roman agricultural writers Cato and Columella. The value of guano was recognized by the Peruvian Incas even before the Spaniards conquered their country; in fact it was so highly prized that it was a capital offence to kill young birds on the guano islands. Guano was first introduced into Europe in 1840, when a firm of Lima merchants sent a cargo of Peruvian guano to London. This was not the first use of phosphatic fertilizers in Europe, however; bones were so generally used in England by this time that battlefields on the Continent had been dug up to supply the demand.

In 1840 the Duke of Richmond suggested that the value of bone as a fertilizer lay in its phosphate content and in the same year the German chemist Baron Liebig suggested dissolving bones in sulphuric acid to make their phosphate content more soluble and available to plants. However, in 1842 John B. Lawes patented a process for treating bone ash with sulphuric acid and to him must go the credit for discovering superphosphate fertilizers. In the short space of 20 years the annual production of superphosphate in England had reached 200,000

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*Figure includes latest estimates of tonnages in the Phosphoria Formation - see later section.

tons. Mined phosphate from the Cretaceous coprolite beds of Suffolk and Cambridgeshire was used as the raw material.

Natural Phosphates, Available Phosphates and Superphosphate

The process of superphosphate manufacture renders available to the plant the phosphate of 'natural phosphates' (such as phosphate rock, apatite, bone and dry guano) by making it soluble in water. In natural phosphates much of the phosphate is combined with calcium and is practically insoluble. If natural phosphates are applied directly to the soil, however, particularly if they are first finely ground, the phosphate becomes slowly available through the action of soil acids.

So called 'available phosphates' are produced as by-products of industrial processes (notably the basic slag produced when iron is smelted from phosphatic ores) or by specific processes, such as calcining and defluorination of phosphate rock. These phosphates are only sparingly soluble in water but readily dissolve in weakly acid solutions, such as are present in some soils. Their value to crops is not universally accepted; certainly the phosphate in them is not so readily available to the plant as it is from superphosphate. The strength of these 'available phosphate' fertilizers is expressed as their solubility in a 2 per cent solution of citric acid or neutral ammonium citrate.

In superphosphate manufacture, raw phosphate rock is treated with sulphuric acid and the insoluble tricalcium phosphate of the raw rock is converted to a mixture of soluble monocalcium phosphate and gypsum:

$$Ca_3(PO_4)_2 + 2H_2SO_4 + 6H_2O - CaH_2(PO_4)_2 + 2H_2O + 2(CaSO_4.2H_2O)$$

This is the basic reaction, but the chemical composition and the nature of the superphosphate vary with the original rock. Superphosphate contains less phosphate than the original rock (17 per cent to 22 per cent P_2O_5 as against 34 per cent to 38 per cent P_2O_5), but the phosphate is soluble. Triple superphosphate (containing 40 per cent to 48 per cent P_2O_5) is prepared by acidulating phosphate rock with concentrated phosphoric acid instead of sulphuric acid. It is a popular fertilizer in America, where its low bulk cuts down freight costs. It has not, I believe, been manufactured so far in this country.

The relative value of natural phosphates, available phosphates and superphosphates as fertilizers is a matter of opinion. Certainly the immediate spectacular increases in plant growth and crop yield associated with the use of superphosphate are not generally found with the slower acting fertilizers. However, some maintain that, since plants draw their sustenance from a soil solution which is relatively dilute with respect to plant nutrients, highly soluble fertilizers may be neither necessary nor desirable. Moreover, proportionately more phosphate is lost by fixation when a water-soluble phosphate is applied to a soil. (Fixation is a process by which soil constituents, notably free iron oxides and alumina, form insoluble compounds with the phosphate). Soluble phosphates suddenly produce a strong solution of phosphate, much of which is washed through the soil and fixed by precipitation in lower soil horizons. Sparingly soluble phosphates on the other hand release P_2O_5 at a rate more nearly conforming to the rate at which the plant can use it, so that proportionately less is washed through, precipitated and lost to the plant.

Chemical Composition of Phosphate Rock

In most phosphate rock the phosphate is chemically combined with calcium and to a lesser extent with iron and aluminium. The superphosphate manufacturer objects to rock with a high iron and aluminium content for two reasons :

(1) These impurities give rise to hygroscopic products which clog seed drills when the superphosphate is spread.

(2) They cause reversion of the phosphate to an insoluble form (C.S.I.R.O., 1943).

There is, however, considerable difference of opinion as to the maximum amount of iron and aluminium that can be successfully handled in rock for superphosphate manufacture. Waggaman (1952) quotes the opinion of various authorities as ranging from 2 to 5 per cent total iron oxides and alumina. Pebble phosphate and rock phosphate from Florida has between 23 and 36 per cent P_2O_5 and between 1 and 4 per cent iron oxides and alumina, but is generally sold with a guarantee of not more than 3 per cent iron oxides and alumina. In the 'black rock' phosphates of Arkansas the percentage of iron oxides and alumina is as high as 9.5 and in the 'brown rock' and 'blue rock' phosphates of Tennessee, upper limits of 8.0 per cent and 7.0 per cent are reported (Waggaman, 1952), although Tennessee rock is apparently not sold with higher than 6 per cent iron oxides and alumina. It is generally agreed that increased care and experience are necessary to process successfully rock with high iron oxide and alumina content. Phosphate rock from Ocean Island and Nauru generally contains less than 1 per cent total iron oxides and alumina, and generally the maximum considered possible for superphosphate manufacture in this country is 3 per cent, although during the war rock with up to 6 per cent of these impurities was accepted.

Another common impurity in phosphate rock is calcium carbonate. When calcium carbonate forms about 5 per cent of the rock, the carbon dioxide formed during acidulation has a desirable leavening effect on the superphosphate formed. If more than 5 per cent calcium carbonate is present, acid is wasted in reaction with it.

Natural phosphates also contain significant amounts of fluorine; for example Florida phosphate has between 0 and 4.4 per cent CaF_2; Ocean Island phosphate has 1 per cent CaF_2; Christmas Island phosphate 3.44 per cent CaF_2, and Morocco phosphate from 5.2 to 9.4 per cent CaF_2. The amount of fluorine present appears to control the solubility of phosphate in natural phosphates and some processes are in operation or proposed, which produce available phosphate by de-fluorination of natural phosphates.

TYPES OF CONTINENTAL PHOSPHATE DEPOSITS

Phosphorite, phosphate rock, or rock phosphate is a sedimentary rock composed mainly of phosphate minerals. A phosphatic rock or a phosphatic formation on the other hand, is a rock or formation in which phosphate minerals form a notable though subordinate part. (Pettijohn, 1949).

Phosphate deposits occurring on continents may be of three types :

(1) Primary bedded phosphorites;

(2) Secondary phosphorites;

(3) Phosphate deposits of igneous origin.

The primary phosphorites are commonly thin beds of great areal extent, interbedded with other marine sediments. The secondary phosphorites are produced by secondary enrichment when a phosphatic sediment, often containing phosphate minerals in only minor amount, is weathered. They are commonly found in place on the surface of the eroding formation, but they may be partly or wholly transported and they may also be moved in solution and redeposited. Deposits of igneous origin are commonly segregations of apatite in large basic alkaline intrusions.

Each of these types of deposit will be examined in some detail, mostly through the description of a well known example of each type.

1. PRIMARY BEDDED PHOSPHORITES

Some phosphate deposits have formed as part of marine sequences. Such deposits are known from many countries of the world and in rocks of many ages; for example in the U.S.A., North Africa, Britain, Belgium, France, Spain, Russia, Brazil, Alaska, South Africa, Poland and Mexico.

Permian Phosphorites of the Western United States

The best known deposits of this type in the world are the Permian phosphorites of Montana, Idaho, Wyoming, Utah and Nevada. The Phosphoria Formation and its partial stratigraphic equivalents cover 135,000 square miles (See Fig. 1 - from McKelvey et al, 1953), The Formation and its equivalents show two distinct facies; in the western part of the area they are part of the strongly folded Cordilleran miogeosyncline and consist of black phosphorites, phosphatic mudstones, cherty mudstones and chert; but in the eastern part of the area they lie on the structurally simple platform bordering the miogeosyncline and are composed mainly of carbonate rocks and a few interbedded cherty, sandy, glauconitic and weakly phosphatic layers. The approximate boundaries of the two facies are shown in Fig. 1; further to the east the Formation and its equivalents tongue into continental red beds. The relationships between the different members and their relative thicknesses are shown in Fig. 2 (adapted from McKelvey et al, 1958). Two phosphatic shale members are now recognized; both reach their maximum development in the Montpelier Canyon area of south eastern Idaho close to the boundary between the two facies.

The main, lower, phosphatic shale member is up to 150 feet thick and contains some 20 phosphorite beds, none individually more than 7 feet thick and together having an aggregate thickness of only 28 feet. However, the beds are persistant and they contain enormous tonnages of phosphate. Recent estimates of reserves in Montana and Wyoming indicate 14,000 million tons of phosphorite with an average grade of 24 per cent P_2O_5 in units less than three feet thick, and a further 21,000 million tons with an average grade of more than 18 per cent P_2O_5 in units greater than three feet thick. (Swanson 1960 and Sheldon 1960).

The phosphorites are composed chiefly of colloform carbonate-fluorapatite, quartz and clay. In addition to phosphorus and fluorine they contain significant amounts of several metals, notably uranium, vanadium, chromium, zinc and the rare earths.

Conflicting theories have been advanced to explain the phosphorites of the Phosphoria Formation. Mansfield (1920), who did much of the earlier descriptive work on the Formation, advanced what has been termed the biolith-diagenetic theory in which the essential process is one of replacement. He thought that first an oolitic limy deposit had formed on the sea floor and that this had later been replaced by the phosphorus of sea water. In later papers (e.g., Mansfield, 1940), he emphasised the role of fluorine in rendering the phosphate insoluble and suggested that periods of phosphorite formation were linked with periods of vulcanism, during which fluorine would be freely supplied to the waters of the basin.*

The alternative theory, that of direct precipitation of the phosphate from sea water, has been developed by McKelvey and others from the work of Kazakov (1937, 1950 - quoted McKelvey et al 1958). Kazakov suggested that where the deep waters of the ocean basins, which are rich in phosphate, well up onto the continental slopes, the phosphate they contain may be precipitated by an increase in the pH of the ascending waters as the temperature increases and the partial pressure of carbon dioxide decreases. Kavakov suggested that, in such ascending waters, calcium carbonate would reach saturation point first and would begin to precipitate and that this would be followed by simultaneous precipitation of calcium carbonate and calcium phosphate after the latter's saturation point had been reached. He suggested upper and lower limits of such precipitation of calcium phosphate as 50 and 200 metres;

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* Evidence of phosphatic replacement on the modern sea floor has recently been found in a sample of phosphatized wood dredged during a survey by the Scripps Institution from a 410 metre terrace in the Gulf of Tehuantepec (S.W. coast of Mexico). The wood is a dicotyledon and is thought to be more than 28,000 years old (C^{14} date). The wood was found partly embeded in clay and only the protruding part had been phosphatized (Goldberg and Parker, 1960).

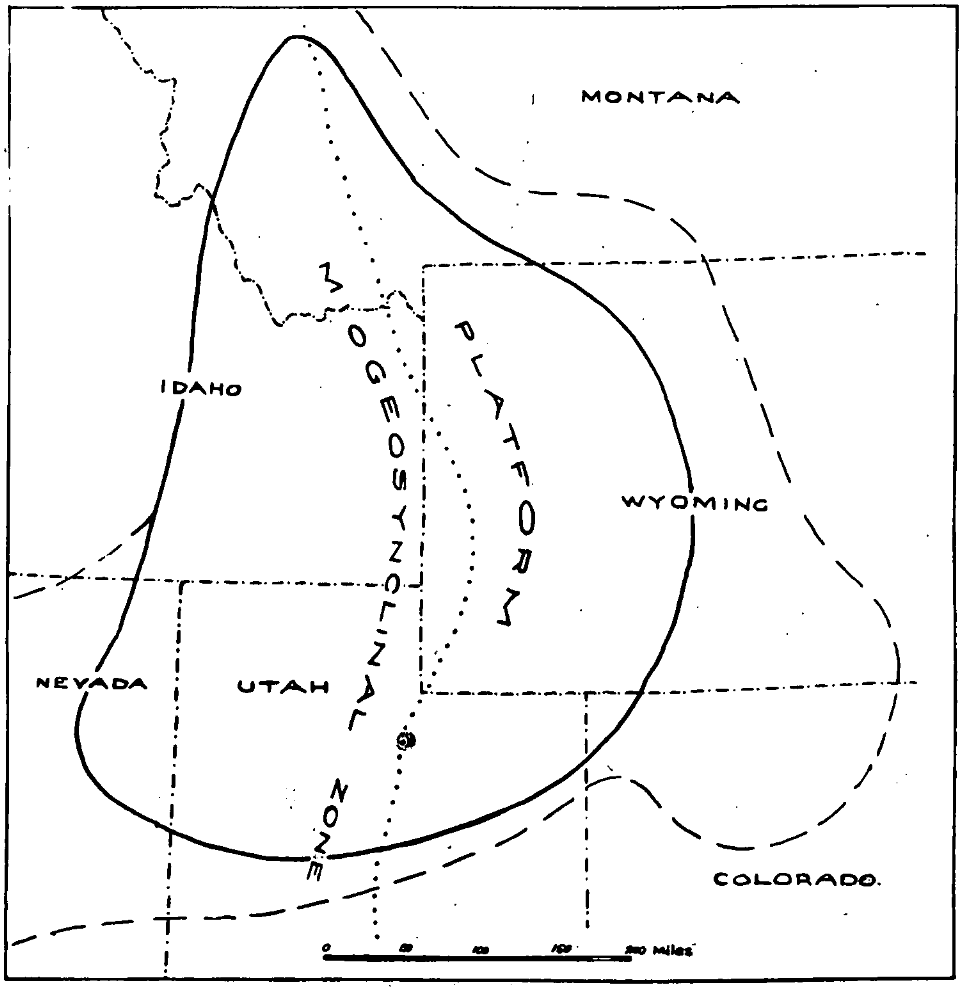

Fig. 1. *Limits of the PHOSPHORIA FORMATION and its partial stratigraphic equivalents (dashed line) and their phosphorite deposits (solid line). From McKelvey et al, 1953.*

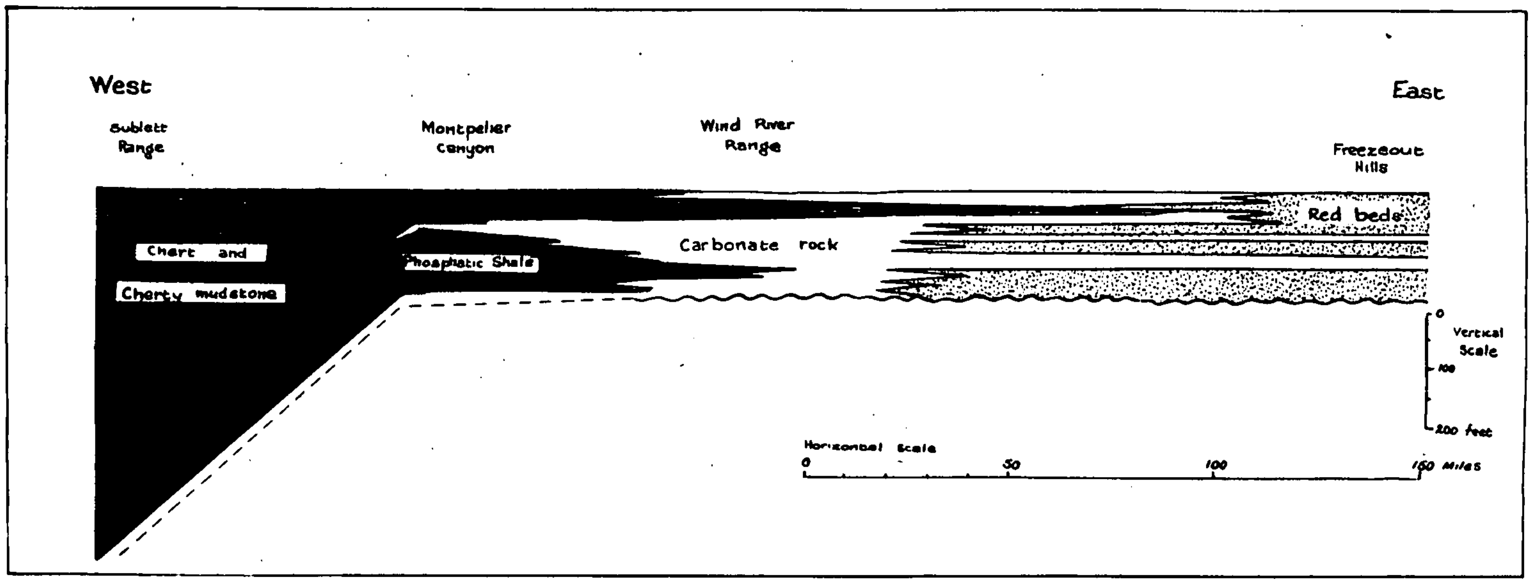

Fig. 2. *Approximate section, Phosphoria Formation (and equivalents) in southern Idaho and Wyoming. (Adapted from McKelvey et al, 1958)*

below 200 metres the waters would not be saturated with regard to calcium phosphate and at less than 50 metres most of the available phosphorus would be used by the plankton population.

McKelvey (McKelvey et al, 1953) has proposed four important modifications to Kazakov's hypothesis. First, the solubility curves of apatite and calcium carbonate overlap but do not coincide completely, so that either may be precipitated independently of the other. Second, that apatite is generally precipitated at lower pH and temperature than calcium carbonate, the reverse of Kazakov's proposition. Third, phosphorites may not be completely the result of chemical precipitation, but may in part be derived from the decay of organisms; since the phosphate of organisms dying near the surface and sinking through water essentially saturated in phosphate would not be resorbed, but would accumulate on the bottom. And, fourthly , the depth at which phosphate is being deposited on the sea bottom at present (Dietz, Emery and Shepard, 1942) suggests that the upper and lower limits of the zone of phosphorite formation may be 200 and 1000 metres, rather than the shallower depths envisaged by Kazakov.

McKelvey also found a close correspondence in areal distribution between phosphorite and chert in the Phosphoria Formation, although the full significance of this is not yet clear.

Calcium phosphate probably accumulates only slowly on the sea floor and phosphorites, of this type, form only where clastic sedimentation is proceeding slowly and where the formation of other chemical precipitates, notably calcium carbonate, is inhibited by the physico-chemical environment. McKelvey envisages the Phosphoria Formation accumulating in a large, shelving basin, bordered by lands of low relief contributing little detritus to the sea. The cold phosphate-rich waters welled up into this basin from an ocean basin to the south or south-west. As the water rose the partial pressure of CO_2 decreased and the pH increased until precipitation of phosphate began. Finally, at even shallower depths and closer to the shore, phosphate precipitation gave place to carbonate.

In later work McKelvey (1959) suggests a fundamental relationship between the occurrence of oil and the occurrence of phosphorite. He says that the up-welling, richly-nutrient coldwaters produce exceptional blooms of phytoplankton, which in turn may produce important accumulations of carbonaceous material in the sediments. The accumulations of carbonaceous matter are the source of oil and, therefore, we can expect both oil and phosphorite to occur in lateral and vertical association with bedded chert, black shale and marine evaporites. He quotes American examples of their association to support his view.

As regards prospecting for phosphorite, he suggests a search in areas of coastal plain sediments adjacent to areas of modern up-welling ocean currents, since he believes that neither ocean currents nor the continental margins have shifted a great deal since the Cretaceous.

The North African Phosphorites

A series of vast phosphorite deposits occur in the Upper Cretaceous and Eocene rocks of the North African countries of Morocco, Algeria, Tunisia and Egypt. In the last few years large related deposits have also been found and are now being worked in the Negev Desert of Israel. The North African deposits are mostly thin beds of incoherent oolitic phosphate in thin sequences of limestone, marl and chert. The phosphorites contain fish and reptile remains and it was first suggested that the deposits had formed as a result of mass mortalities of fish with changes in water circulations in a shallow sea.

Cayeux (1950) studied the North African deposits in considerable detail. He made various deductions as to the environment of deposition but of particular interest is his conclusion that bacteria played a major part in the precipitation of the phosphate. He studied thin sections of the North African phosphate at the largest available magnifications but did not recognise the bacteria until he tried using infra-red illumination.

He then found his slides crowded with small spherules .5 to 2.5 microns in diameter.

Charles (1953) follows Cayeux in stressing the part played by organisms in the precipitation of phosphate. He envisages a *zone of formation*, where algae are active in extracting the phosphate and a *zone of accumulation* at some distance from the zone of formation, where the phosphate accumulates.

Salvan (1953) describes the Moroccan deposits and favours a theory of chemical precipitation similar to that proposed by Kavakov. He tentatively, and rather hesitantly, proposes that the source of the phosphate in the sea water is under-sea vulcanism.

A striking feature of the North African deposits is the very thin sequence in which they occur. This sequence of at least partly chemically precipitated limestones, marl and chert is without obvious erosion breaks but stretches through the upper Cretaceous and into the Eocene. A fundamental requirement for the accumulation of workable phosphate deposits is that clastic sedimentation should be at a minimum.

Australian Occurrences

The deposits at Phosphate Hill, Howe's Creek, and Whitfield in the Mansfield district of Victoria (Fig. 4) occur with bedded fossiliferous rocks of upper Cambrian age. (Prichard, 1951; Howitt, 1923). The phosphorite beds are associated with radiolarian cherts, fossiliferous slates and shales. The rocks are folded, crumpled and faulted and strata are crossed in all directions by filaments of wavellite, turquoise and quartz. The phosphate is principally aluminous although locally calcium phosphate predominates. The phosphate is unsuitable for superphosphate manufacture because of its low grade and high aluminous content. The percentage of P_2C_5 varies between 1 and 23 per cent - 12,012 tons of ore removed between 1916 and 1920 averaged 16 per cent P_2O_5 with 19 per cent Al_2O_3 and Fe_2O_3.

No reserves are stated, but they would certainly be small. The occurrence does not give any hope of finding larger or more continuous bodies in the same strata elsewhere.

A marine phosphorite deposit also occurs at Dandaragan, Western Australia, about 100 miles north of Perth (Fig. 6). Two beds, each about two feet thick, of phosphate nodules in a matrix of glauconitic chalk (Woodward, 1917). The beds occur over an area 24 miles long by 4 miles wide; but with a 20 foot stripping limit they would yield less than a million tons of low grade phosphate. Also the phosphate is high in iron oxides and alumina, and can not be readily beneficiated.

SECONDARY PHOSPHORITES

During a period of prolonged erosion, an exposed phosphatic limestone may have its phosphate content concentrated on the surface as the carbonate of the limestone is removed by solution. The phosphate forms a blanket of pebbles and small grains lying on the irregular etched surface of the limestone.

THE FLORIDA DEPOSITS

The best known secondary phosphorite deposits in the world are those of Florida. It is generally agreed that these were formed as the phosphatic limestone of the Miocene Hawthorn Formation was eroded, probably during the late Miocene. The undisturbed insoluble residue of phosphatic pellets and nodules, quartz sand and montmorillonitic clay, concentrated on the surface of the Hawthorn Formation or on the underlying Ocala limestone (Eocene), are known as *'land pebble'* phosphates (Fig. 3). They occur under a shallow cover of Pliocene and Pleistocene sand and gravel along much of the central part of peninsular Florida (see Fig.4). 'Land pebble' phosphates that have been transported, and commonly concentrated, by river erosion, are termed *'river pebble'* deposits and spread to the south and west of the

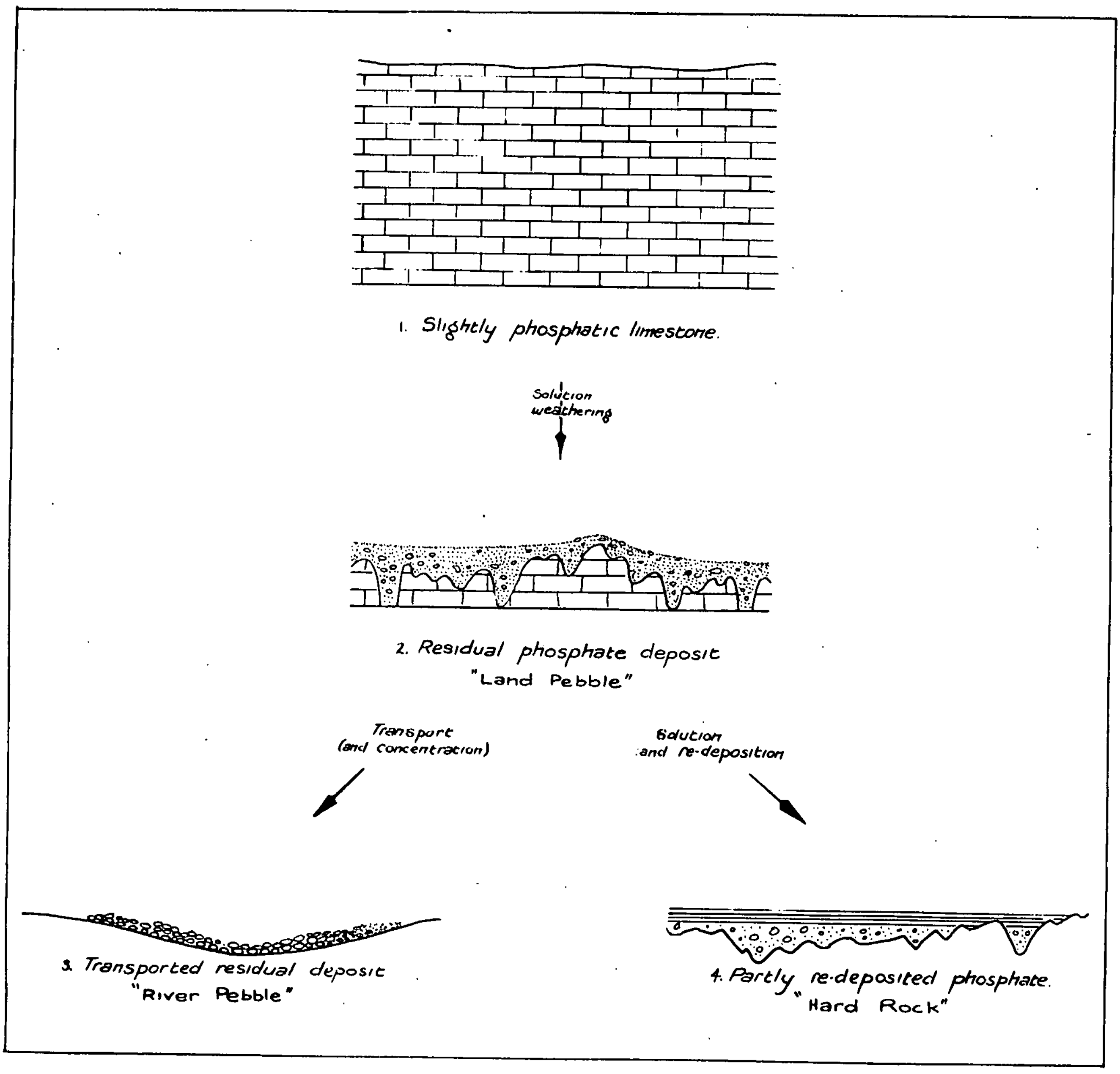

Fig. 3. Formation of secondary phosphorites.

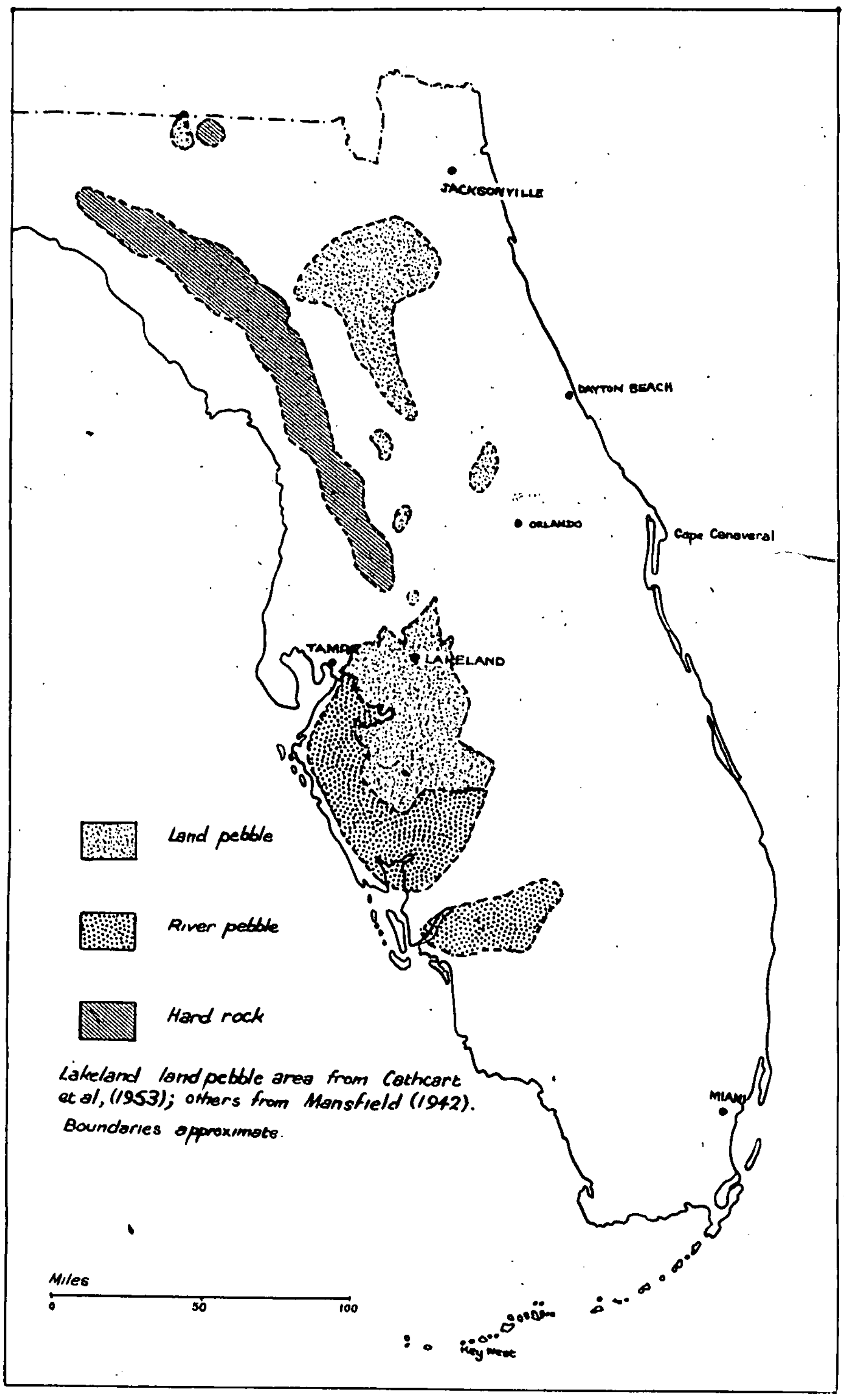

Fig. 4. Approximate boundaries of phosphate deposits, Florida.

'land pebble' areas; 'hard rock' deposits, in which some of the phosphate has been dissolved and re-precipitated, occur in a curving belt following the Gulf coast in the north-west of the State.

The 'residual' theory of origin of the Florida phosphate deposits is confirmed by the fact that minerals present in the underlying limestone persist upwards into the overlying deposits in accordance with their solubilities. The least soluble constituents are most abundant in the higher beds; the uppermost bed consists of quartz sand; the next below is of quartz sand and aluminous phosphates; next below that is sand, clay and apatite, and immediately above the limestone is a 'bed clay' of sand, clay, apatite and carbonate. Another characteristic that points to a residual origin is that cross-sections show that the unconsolidated material of the phosphate deposits conforms generally to the irregularities in the underlying limestone, rather than tending to fill depressions and thin out over highs.

The alternative depositional hypothesis was proposed by Sellards (Sellards 1915a, quoted in Cathcart et al, 1953). He explains the vertical changes in the phosphate deposits by supposing that the lower unit (sand, clay and apatite) was deposited in Pliocene time in a shallow marine environment, the material coming from erosion of the Hawthorn Formation at the end of the Miocene. The upper unit of the phosphate deposits (sand and aluminous phosphates) was, he supposes, laid down in a marine offshore environment. Subsequently, the succession was eroded and surficial terrace sands were formed during the Pleistocene.

The solution and re-deposition of phosphate to form the 'hard rock' deposits appears to take place if the groundwater contains humic acids. Early experiments by Graham (1925) suggest that, where natural waters contain CO_2 , calcium carbonate is dissolved more easily than clacium phosphate (in the ratio 1:100 :: P_2O_5:CaO); but where humic acids are present a considerable quantity of phosphate is also dissolved. As long ago as 1892, Reese suggested that phosphate was dissolved by water containing dissolved organic matter, but if this solution was allowed to stand over calcareous beds, the phosphate re-deposited (Reese, 1892, quoted Sellards 1915b).

'Land pebble' and 'river pebble' deposits occur over some 2,600 square miles of Florida. The measured reserves of the fields in the State are around 2,000 million long tons (Mansfield, 1942). The grade is between 25 percent and 33.5 percent P_2O_5 and, as the deposits are generally of loose gravel and sand under very little overburden, they can be worked cheaply (commonly by sluicing) and the ore can be easily beneficiated by flotation, washing and screening.

Similar deposits occur in South Carolina, North Carolina and in Tennessee. The Tennessee deposits contain much re-deposited phosphate.

Australian Occurrences

Small residual phosphate deposits occur in a 200 mile long meridional belt in South Australia, from Carrieton in the north to Myponga in the south. The deposits appear to have formed during a Tertiary weathering cycle on the surface of Cambrian and Precambrian limestones. The phosphate varies from a massive rock to a chalky powder; breccia, mamillary crusts and cavity fillings are also common. It is mostly an anhydrous lime phosphate, but in some places replacement of shale and slate has produced a predominantly aluminous phosphate.

Reserves in the whole belt do not seem to total more than 120,000 tons and the material is said to be mostly too high in iron oxides and alumina for superphosphate manufacture.

APATITE SEGREGATIONS

The Khibina Tundra

Probably the largest phosphate deposit of igneous origin in the world is that of the Khibina Tundra, Kola Peninsular, U.S.S.R.. Reserves are said to be more than 2,000 million tons (of apatite) and production during 1956 was estimated at 3,690,000 tons.

The apatite of this deposit is segregated in a large basic igneous intrusion. As mined the ore contains about 30 percent P_2O_5 and is chiefly contaminated with nepheline, aegerine, titano-magnetite and sphene. The ore is concentrated by a flotation process to remove these impurities; this concentrate contains 40 percent P_2O_5.

Other deposits of this type occur in Norway, Sweden (Horesjobera), Canada (N. Burgess and at Nemego, Ontario) and in Brazil. Recently a deposit has been discovered at Sukulu, near Tororo, Uganda where there are said to be reserves of 200 million tons with an average grade of 13 percent P2O5. Interestingly enough, this deposit was largely ignored by the geologist who first mapped the area.

Australian Occurrences.

In South Australia, concentrates of apatite in copper-bearing pegmatites have been noted from Moonta and Wallaroo and at Boolcoomata Station, north of Olary, granite pegmatites contain considerable masses of apatite. These have been prospected and found to be insignificant economically. (David, 1950 Vol II, p. 350).

DISTINCTIVE FEATURES AND COMMON ASSOCIATIONS OF PHOSPHATE DEPOSITS

Distinctive Features

Bedded primary phosphates are characteristically black. Oolites and concretionary nodules are nearly always present and in some deposits all the phosphate is contained in them. An imperfect concretionary structure is characteristic, but is not always present. The oolites commonly show a concentric structure only in their outer shells; they never have the inorganic nuclei (sand grains etc.) nor the radial structure, so characteristic of calcareous oolites.

Residual phosphates may be white, yellow or, more rarely, brown and may be composed of clay, sand, and pebble-size material in various proportions. Phosphate 'pebbles' or nodules are commonly irregular in shape, with a mamillated outer surface. Broken surfaces have the typically soapy appearance and unctuous feel of cellophane, and may be black, brown, white or grey. The black colour is commonly most intense near the outer rim of the pebble. Many pebbles are crowded with oolites.

Hydrocarbons are commonly included in phosphorites and produce a fetid odour when the phosphorite is struck.

The high fluorine content of phosphorites commonly leads to high fluorine in groundwaters associated with them. Burkelow (1946) records between 1.0 and 3.4 ppm of fluorine in groundwaters of Sarasota County, Florida, which flow through the miocene Hawthorn Formation (see section on Florida deposits). The fluorine content of water supplies in Algeria, Morocco and Tunisia is high enough to cause fluorosis among the inhabitants.

Recently an unexposed phosphorite deposit in Beaufort County, North Carolina was accidentally located during an underground water survey (Brown, 1958). Groundwater from the phosphorite is significantly higher in halogen content and the limits of the phosphorite can be delineated by a study of the chemical analyses of the groundwater. The phosphorite, tentatively regarded as middle Miocene in age, is buried under 45 to 250 feet of late Miocene marl. The deposit varies from a few feet to a maximum of 90 feet thick; it lies unconformably on a limestone of Eocene age; and is a loose sand containing between 8 and 31 percent P_2O_5. The main diluent is sandsize quartz, but the phosphorite also contains some silt, clay and organic material. The associated rocks are glauconitic sand, argillaceous sand and impure limestone.

Geochemical enrichment of primary phosphorites in uranium, along with other metals such as vanadium, chromium, zinc and the rare earths, has already been noted in the section on the Phosphoria Formation. The radioactivity associated with this enrichment of phosphorites in uranium may serve as an aid to the recognition of phosphorite horizons, particularly by the examination of gamma ray logs of drill holes. (See "Wilgunya Formation" in Recommendations Section).

Mineralogy

The minerals present in phosphorites are often conveniently referred to by the general name 'collophane'. The collophane of a number of phosphorites has been shown to be a crypto-crystalline member of the apatite group, close to fluor-apatite ($Ca_6F_2(PO_4)_6Ca_4$). In the Florida phosphorites, the dominant mineral is commonly francolite, which is a carbonate apatite with fluorine, and dahllite, which is a carbonate apatite with hydroxyl (McConnell, 1950, 1958). Where the phosphorite contains aluminium, it is probably present at the mineral crandallite ($CaAl_3(PO_4)_2(OH)_5.H_2O$).

Collophane is opaline in appearance, with a dense, layered or colloform structure, and greyish white, yellowish or brown in colour. Dana (1957) gives the hardness as between 3 and 4, but in my experience this hardness is commonly exceeded.

Chemical Spot Test for Phosphate.

The best test for collophane or any of the phosphate minerals is a spot test for the phosphate radical. The following test (Hicks, 1927) is sensitive and will detect phosphate in small quantities:

(1) Moisten fresh rock surface with a drop of nitric acid (S.G. 1.2)

(2) Place a small crystal of ammonium molybdate on the spot.

(3) If phosphate is present a yellow colour (ammonium phospho-molybdate) gradually spreads throughout the crystal and over the rock surface.

The same test may be made by crushing a little of the rock and placing it in a test tube with a few drops of nitric acid (S.G. 1.2), diluting with a little water and adding ammonium molybdate solution in excess. If phosphate is present, ammonium phospho-molybdate forms a yellow precipitate.

To a limited extent the intensity of the yellow colour or the volume of the precipitate may be used as a guide to the amount of phosphate present.

COMMON ASSOCIATIONS OF PHOSPHATE DEPOSITS

Three things which, either together or separately, are commonly associated with bedded phosphorites and have some significance in prospecting, will be mentioned:

(1) Glauconite and greensand;

(2) Surfaces of unconformity or disconformity;

(3) Thin sequences of thin bedded, chemically precipitated sediments without intervening clastics.

(1) Several authors have noted the association of marine phosphorites with glauconite and greensand. Goldman (1922) points out the association between the two and relates the occurrence of both to unconformities or periods of non-deposition of clastic sediments. Glauconite is associated with the Tennessee phosphorites, with beds of phosphate nodules in the Cretaceous of England (Fisher, 1873, quoted by Pettijohn, 1949), with the sandy phosphorites of the ~~Upper~~ Cretaceous of Russia (Bushinsky, 1935) and, to a limited extent, with the platform facies of the Phosphoria Formation. Teall (1900) records glauconite in the phosphate deposits of North Africa. In the deposits of the Eocene of Dahomey, on the other side of the Sahara, the association of glauconite and phosphorite is strongly marked (Slansky, Camez and Millot, 1959). In an interesting new find in North Carolina (Brown 1958), the phosphorite is associated with glauconitic sands.

(2) Grabau (1919) has pointed out the important relationship between bedded phosphate deposits and surfaces of disconformity. He explained the phosphorite occurring at such horizons as a concentration

of phosphatic material that had originally been scattered through a formation now missing (see "Secondary Phosphorites"). He also notes that the common bedrock of phosphate deposits is limestone.

The Phosphorite beds of the Miogeosynclinal Zone of the Phosphoria Formation are associated with black shale, chert and limestone. The phosphorite in this type of deposit occurs in a thin sequence with no coarse clastics associated with fine bedded chemically precipitated limestone and chert. The whole sequence is thin and individual beds within the sequence are thin but continuous over long distances.

In North Alaska, a recently discovered bedded phosphorite occurs in a very similar sequence in the black shale and chert member of a limestone of Mississipian age (Patton and Matzko, 1959).

The vast deposits of Morocco, Algeria and Tunisia are similarly of thin beds in thin sequences mainly of marl, limestone and chert, In the Khonribga deposit (Morocco) the sequence (Maestrichtian to the base of the Lutetian) is only 25 metres thick and contains 6 phosphorite beds (Salvan, 1952). The main phosphorite bed has an average thickness of 1.7 metres and is seldom more than 2.7 metres thick (Lawson, 1931).

Chert is associated with the Upper Cretaceous phosphorites of the Negev Desert, Israel, which have only recently been discovered and are now being worked (Hoffman and Mariacher, 1961 and Bentor, 1954).

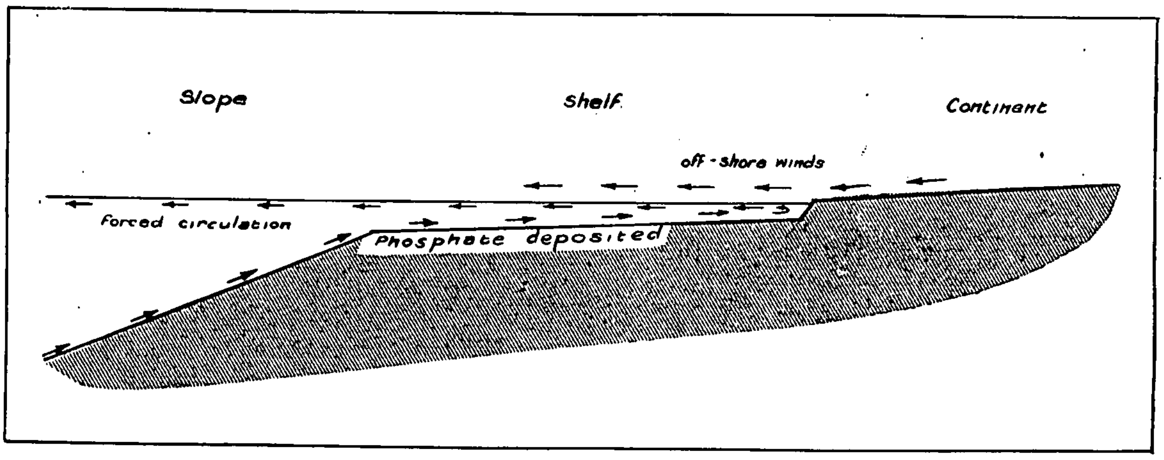

Fig. 5. Deposition of phosphate on continental shelf

UNDERSEA PHOSPHATE DEPOSITS

Phosphate nodules are forming at a number of places on the sea and ocean floors. The nodules vary in size from oolites to boulders more than three feet in diameter and they seem generally to form a single layer over the sea-floor where they are found. In environments where little detrital material is being added important concentrations of these nodules exist.

The best known occurrences of this kind are those off the coast of California. Over the last few years, largely because of research undertaken by the Scripps Insititution of Oceanography, so much bottom sampling has been done in the area that it is now possible to delineate accurately those areas over which concentrations of phosphate nodules occur. Recently interest has been aroused in the possibility of mining these deposits. Mero (1959, 1960 and pers.com.) has examined the economics of mining them by drag dredging and by hydraulic dredging. Interest centres on one deposit, the Forty Mile Bank deposit (40 miles west of San Diego). The phosphorite averages about 29% P_2O_5 and reserves are thought to be about 3.1 million tons in water averaging 600 feet deep. Mero concludes that it would be possible to dredge this deposit profitably provided that a market for 40,000 tons of phospherite per annum can be found.

These deposits of phosphate nodules on the continental shelf are thought to have formed by a similar mechanism to that proposed by Kavakov and later McKelvey et al for the bedded phosphorites (See Section on Phosphoria Formation). The process is illustrated in Fig. 5. Off-shore winds from a continental margin cause a circulation of water in a vertical plane with cold water from the deeper ocean welling up to take the place of the warmer surface water, which is moved by the wind out to sea. The cold bottom water, which is rich in phosphate, rises over the continental slope and on to the continental shelf depositing its phosphate as it loses CO_2 and its pH changes.

Some geologists, however, explain the off-shore Californian occurrences as residual deposits formed by some process of under-sea erosion or solution of a phosphatic limestone.

In contrast to the information available about the bottom deposits of off-shore California, the amount of information available about Australia's continental shelf is meagre. Mero (1960) notes that phosphate nodules were dredged from the continental shelf off the east coast of Australia by scientists of the Challenger Expedition in 1874. I have not been able to confirm this report or to find a more precise description of the locality. In the "Deep Sea Deposits" volume of the Challenger Reports (Murray & Renard, 1891), the authors state: "During the Challenger Expedition phosphate of lime was produced at many of the shallower stations around continental shores" (p.396). At eight stations 'off Sydney' in a triangle between 33°57'S, 151°39'E, 34°13'S, 151°38'E and 34°8'S, 152°0'E (Stations 163D to 164B), green sand and green mud are reported. This is the same association as that of the phosphate nodules of the Agulhas Bank, off the Cape of Good Hope, but phosphate nodules are not definitely noted for the Australian stations.

There are two basic lines of approach to the problem of locating accumulations of phosphate nodules on the continental shelf; one is to prospect systematically over the shelf by dredging samples, the other is to study water movements to locate vertical circulations of the type described earlier and shown in Fig. 5.

With regard to the first, we can expect that much more information will become available during the next few years from bottom sampling on the shelf. At present, with the exception of the 1960 Scripps Institution work in the Timor Sea, virtually nothing is known from dredging. The 'Challenger' only crossed the Australian continental shelf in a few places - in Bass Strait, along the east coast as far north as Sydney, and again on the east coast near Cape York.

In modern surveys, the information laboriously obtained at a few fixed stations by dredging can be augmented by under-sea photography between the stations. The indications shown on Admiralty charts are from material adhering to the greased base of the sounding lead and have very little significance.

Water movements off the Australian coasts have only been studied in detail in a few places. The Marine Biology Laboratory of the Commonwealth Scientific and Industrial Research Organization's Division of Fisheries and Oceanography at Cronulla, New South Wales, is constantly engaged in research in this and in allied fields, but Australia has a long coastline and a prodigious amount remains to be investigated. In at least one instance, however, a vertical circulation similar to that of Fig. 5 has been noted. During four winter months (May to August), consistent off-shore winds and off-shore water movements have been recorded between 1953-1956, by drift card releases from eight stations between Evan's Head and Eden along the New South Wales Coast (Chau, 1957). Ovservations on the variation with depth, of water temperature, phosphate content, and oxygen content, were carried out at stations in line at right angles to the coast. These confirm that a vertical circulation of water is occurring, with upwelling cold water taking the place of surface water forced out from the shore by winds.

The continental shelf in this area should certainly be sampled by dredging at the earliest opportunity.

RECOMMENDATIONS

General

The main purpose in preparing this report has been to draw attention to Australia's acute need of new phosphate reserves and to present information on the appearance and common association of phosphorites which may assist in finding deposits on the mainland.

Three points should be stressed :

(1) The very large tonnages that can be contained in very thin beds.

(2) The great economic value of bedded phosphorites.

(3) The difficulty of recognizing phosphorite.

(1) The tonnages contained in bedded phosphorites are commonly very large. Roughly a bed 3 feet thick extending for one square mile would contain about 4 million tons of phosphorite. Primary bedded phosphorites are characteristically thin beds of great areal extent; therefore, any occurrence of phosphatic material, however inconspicuous, is significant and should not be dismissed without careful appraisal.

(2) The great economic value of phosphorites is inherent in the fact that such thin beds can contain such vast tonnages. The value of phosphorites should be brought to the notice of prospectors, along with information of the kind contained in this report to help in identification.

(3) Phosphorite is not easily recognised; it is very easily mistaken for limestone. One island deposit was described by a geologist, who had very considerable experience on coral islands, as a silicified oolitic limestone, even after thin sections of the material had been made and although he was at a loss to explain the silicification. Any dense, dark or black limestone, particularly if it has an oolitic structure, is worth testing chemically.

Specific Targets

Known Deposits

There seems to be very little point in revisiting the known occurrences of phosphorite on the continent. The extent of each deposit appears to be established and none seem likely to be more extensive or to re-appear elsewhere. Recently the Kapunda - Robertstown area, South Australia, where small phosphorite deposits are known, was prospected thoroughly for asbestos and phosphorite, but no important finds were made (Forbes, 1959).

The known deposits are summarized in Table II and the localities are shown in Fig. 6.

ports of phosphatic horizons

Two formations from which phosphatic material has been reported should be investigated further:-

(1) The Wilgunya Formation

(2) The Bulgadoo Shale

(1) The Wilgunya Formation

During 1959, a Bureau of Mineral Resources field party mapped a poorly outcropping fine-grained rock in the Boulia area, which was later found to be quite strongly phosphatic (Casey et al, 1960). The outcropping rock is thought to be part of the Wilgunya Formation, a formation dominantly of dark shale, siltstone, calcareous siltstone and limestone, in the Lower Cretaceous sequence of the Great Artesian Basin. The Wilgunya Formation contains the Toolebuc Member, 30 feet of fossiliferous limestone and calcareous siltstone, which has been recognized in a number of bores because of a gamma-ray count which is 3 to 4 times normal background count (Reynolds et al, 1961.) The association of high radioactivity with phosphorite beds has already been noted. Also interesting is the fact that the Toolebuc Member changes lithologically towards the south and becomes a glauconitic limestone.

The Wilgunya Formation should be examined in outcrop and in core and cuttings from completed drilling. Any proposal for drilling likely to penetrate the Wilgunya Formation should take into account the significance of the formation as a possible source of phosphate.

(2) The Bulgadoo Shale

The Bulgadoo Shale is a formation in the Byro Group in the Lower Permian (Artinskian) of the central area of the Carnarvon Basin, Western Australia. The Bulgadoo Shale consists dominantly of carbonaceous shale and siltstone with minor quartz greywacke and limestone beds (Condon 1954). It contains a number of thin beds of probably phosphatic siltstone, which characteristically show a reticulating pattern of one lithology in another dominant lithology, for which Condon proposes the name "network beds". The unusual form of these "network beds" is thought to be due to shrinkage, but no definite evidence has been found in the deeply weathered outcrops so far examined. Simpson (1926, p.93) records 7.35 per cent, P_2O_5 in a specimen, which was from an outcrop of the Bulgadoo Shale on the Minilya River west of Wandagee Homestead.

The Bulgadoo Shale should be examined in outcrop and in core and cuttings from drilling. Phosphate estimations should be made on specimens selected systematically across the thickness of the shale.

Localities with High Fluorine Content in the Groundwater

The apatite mineral of phosphorites is commonly close to fluor-apatite and phosphorites are commonly associated with high fluorine concentrations in groundwater (See Section : Common Association and Distinctive Features).

The medical and dental professions are interested in the fluorine content of water supplies, since the prolonged drinking of water with high fluorine content produced dental abnormalities ('mottled enamel' and malformation of teeth) and, in severe cases of fluorine poisoning, osteosclerosis. On the other hand, the presence of a certain amount (about 1 ppm) of fluorine in the drinking water is believed to be advantageous, particularly to growing children, in preventing dental caries and the addition of fluorine to town water supplies to achieve this level has been advocated.

As a result of this interest, a certain amount of information has been collected on the amount of fluorine in groundwaters in Australia. The available information is from scattered localities only, but does indicate a number of places with particularly high concentrations of fluorine in the groundwater. Localities with greater than 1 ppm of fluorine are shown in the the accompanying table (Table 3). For comparison it should be noted that groundwater of Sarasota County, Florida, has between 1.0 snd 3.4 ppm of fluorine (Burkalow, 1946). It is not possible to give an average figure for groundwater as the available data relate to 'domestic water supplies', whether groundwater or surface water.

Ward (1954), who records the high fluorine content of water from bores near Maree leaves open the question of a source for the fluorine. He considers, but discounts, the possibility of a magnetic or volcanic source and he mentions the possibility that the fluorine is derived from phosphate beds, but notes: "So far as is now known, there are no phosphate horizons in the water-bearing beds".

The high fluorine water is described by Ward as "carbonated water of eastern origin in the principal aquifers of the Great Artesian Basin".

By contrast the water from eight bores, intersecting aquifers carrying water of western origin, along the western edge of the Basin in South Australia average only 0.6 ppm of fluorine.

The occurrences of high fluorine in groundwater near Maree should be examined carefully; in particular any new boring undertaken there should be logged by a geologist on the spot and cuttings should be chemically tested for phosphate. The first step is to identify a phosphate horizon in the succession, if one is present, and then to consider where the horizon is likely to crop out at the surface.

The other localities listed in Table 3 are also of interest. The Great Artesian Basin localities in New South Wales are also towards the edge of the Basin and may have a related origin to the Maree bores. The localities in Western Australia may be related to a Permian succession containing the Bulgadoo shale.

Table 3. Localities with high fluorine in groundwater.

Basin	State	Locality	Fluorine (p.p.m)	Reference
Great Artesian	S.A.	25 Bores near to Maree (average)	4.1	Ward (1954)
		highest	13.5	
Great Artesian	Qld.	Quilpie	1.5	Dept. of Health File 100/3/1
		Thargomindah	3.0	
Great Artesian	N.S.W.	Carinda	1.15	Jones (1949)
		Moree	2.20	
		Warren	1.25	
Murray	Vic.	(Kaniva	1.0)	Dept. of Health File 100/3/1
		(Nhill	1.0)	
	S.A.	(Bordertown	1.1	Ward (1954)
		(Naracoorte	1.2	
Unnamed (Tertiary)	S.A.	Wanilla	1.3	Dept. of Health File 100/3/1
Carnarvon	W.A.	Area north of Geraldton	High *	Dept. of Health File 100/3/1

* "Toxic concentrations in isolated localities".

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TABLE I : SUMMARY TABLE OF CONTINENTAL PHOSPHATE DEPOSITS

Type of Deposit	Common Association	Distinctive Features
(1) PRIMARY (a) Platform	Glauconite, greensand, limestone	Thin sequences, oolitic structure, marine transgression. Dark colour
(b) Geosynclinal	Chert, black carbonaceous shale and smaller amounts of limestone	Thin sequences; thin bedded black shale, lack of clastics. Oolitic structure. Dark colour.
(2) SECONDARY	Underlain by limestone, contains clay, sand unconformity	Oolitic structure; mamillary crusts of collophane. Pebbles of cemented oolites. Uneven thickness of deposit.
(3) IGNEOUS	Nepheline Syenite intrusions; nepheline, aegerine, magnetite, sphene etc.	

TABLE II. PHOSPHATE DEPOSITS ON MAINLAND AUSTRALIA

Deposit	State	Locality	Occurrence	Associated Beds	Age	Previous Production				Estimated Reserves in tons	References
						Years	Tonnage	% P_2O_5	% $A6_2O_3$ $+Fe_2O_3$		
PRIMARY											
Phosphate Hill	Vic.	Near Mansfield	Lenticular bodies in crumpled strata. Bedded fossiliferous phosphate rock.	Chert, slate, shale	Upper Cambrian	1916-20 1921-26	12,012	16	19	No tonnage stated Grade between 1 & 23% P_2O_5	Howitt (1923)
Howe's Creek	-	-	Related but smaller deposit			Unworked					
Dandaragan	W.A.	100 miles N. of Perth	Lower phosphate bed: Ph. nodules and fragments of wood replaced by apatite in greensand overlain by glanconitic sandstone and underlain by ferruginous sst. Upper phosphate bed: Ph. nodules in glaunconitic chalk. Overlain by chalk; underlain by greensand.	ferruginous sst., glauconitic sst., chalk, greensand	Cretaceous		NIL			691,000 of 6% P_2O_5 (High in Fe and Al.) 263,000 of 11% P_2O_5 Equivalent: 954,000 of 7.5% P_2O_5	Woodward (1917) Campbell (1907)
SECONDARY											
Orroroo	S.A.	Deposits are in a belt from Carrieton in the North to Myponga in the South.	Erosion of Cambrian and Upper Precambrian limestones during a Tertiary weathering cycle	limestone	Tertiary	1943/4	1,210			-	Dickinson (1943)
Clinton	"					-	-			-	
Tom's	"					to 1919	20,000	18.7 to 19.5	23	20,000	
St. Kitt's	"					to 1919	20,000			-	
						)1919-22)1943-44	4,349	20 to 22		76,000 (20% P_2O_5 10% oxides)	
Koonunga	"					to 1919	3,000			-	
Klemm's	"					-	-			22,200 (18.5% P_2O_5 - High oxides)	
Lower Hermitage	"					1941,43,44	862	Good.		-	
Myponga	"					-	-			-	
IGNEOUS											
Alcoota Stn.	N.T.		Apatite and magnetite at limestone/pegmatite contact.							Negligible.	
Boolcoomata Stn.	S.A.	N. of Olary	Granite pegmatites contain considerable masses of apatite							Prospected but found uneconomic	
Moonta	"	Near Wallaroo	Apatite occurs in copper bearing pegmatites							-	

Fig. 6

AUSTRALIA & NEW GUINEA
SEDIMENTARY BASINS
with
Localities Mentioned
in
RECORDS 1961/120

Bureau of Mineral Resources, Geology and Geophysics.