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The amygdale minerals in the Tertiary lavas of Ireland. III. Regional distribution.

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Summary.—Zeolites occur at most of the 670 localities studied in the Antrim Basalts, and the ten most abundant species, in order of frequency, are chabazite, thomsonite, analcime, levyne, natrolite, mesolite, stilbite, gmelinite, heulandite, and phillipsite. Gismondine, laumontite, and mordenite are recorded for the first time. Chabazite and thomsonite characterize the upper parts of the lava succession. Below, they are joined by analcime and natrolite and, in some areas where zeolitization is most intense, by stilbite and heulandite as well. These mineral zones have been mapped, and they probably reflect the temperature distribution in the lavas during zeolitization. Being discordant and clearly superimposed upon the lavas, these zones establish the late age of zeolitization in the volcanic history of Antrim. In places the zeolite zones are missing, and in their place a mineral assemblage rich in calcite, aragonite, quartz, and chalcedony is found. The tholeiite lavas that occupy a small area in north Antrim have an amygdale mineral assemblage different from that in the olivine-basalts, which constitute the remainder of the lava outcrop.

THIS paper is the third of a series dealing with the distribution of zeolites and other secondary minerals in amygdales in the Antrim Basalts. The study has been based on the examination of these minerals at some 670 localities distributed over most of the area of the basalts. The first two papers dealt with certain aspects of the distribution of the minerals; one (Walker, 1951) described the mineral zoning and distribution of chabazite crystal habit in a small area of well-exposed lavas in the east of Antrim, and the other (Walker, 1959*a*) was concerned with the peculiar distribution of gmelinite. In the present paper the regional distribution of minerals in the whole of the Antrim Basalts is discussed, and the two areas already described are located in their regional setting.

The zeolites in the Antrim Basalts have long been well known, owing largely to the work of the early nineteenth-century mineralogists.

The early work consisted mainly in the collection and description of minerals from a limited number of notable localities, and as such was reasonably complete and reliable, although the earlier workers were handicapped in their determination of the fibrous or acicular zeolites by the inadequacies of the methods then available (Hey, 1933). The work was continued in the first decades of this century by the late Robert Bell of Belfast and F. N. Ashcroft, who made extensive collections mostly from the country near Belfast. These collections are now housed in the British Museum (Natural History).

The present study was undertaken in the first place because it was suspected that the minerals were distributed in mappable zones, and it was felt that if this were so knowledge of the pattern of their distribution might help solve the difficult problem of the age and origin of zeolites in basaltic rocks. In the event, these hopes have been justified.

The amygdale minerals and their relative abundance.

The list below gives the zeolites that have been identified by the writer in the Antrim Basalts. It includes some other secondary minerals, but does not include various cryptocrystalline or amorphous substances (e.g. saponite, celadonite), which are locally abundant but which have not been studied in the present investigation. Also omitted are augite and ilmenite crystals, which commonly project into amygdales; the andradite, ilmenite, apatite, and other minerals found at Portmuck, Islandmagee (Walker, 1959*a*); and the copper minerals, which are occasionally encountered in minute quantities. Crystal cavities similar to those in the Watchung basalts, which are ascribed to anhydrite (Schaller, 1932), have been seen at one locality.

It is not easy to arrive at a reliable objective estimate of the relative abundance of the minerals. The method used here is to determine the percentage of localities at which each mineral has been identified, of the 670 localities visited in this investigation; even these figures are not fully representative, on account of the unduly large concentration of localities in some parts of the Antrim Basalts (fig. 4) where exposures are numerous, and no allowance has been made for the different relative abundance of the minerals at each locality. However it is believed that Table I gives a reasonably reliable picture of the relative abundance.

Levyne, although not usually regarded as an important mineral, is revealed as one of the more widespread of the zeolites although, on account of the usually small size of the crystals, it is rarely conspicuous. Gismondine, laumontite, and mordenite are new records for Antrim, while the writer considers that the rather doubtful records by earlier workers of epistilbite, brewsterite, and harmotome cannot be substantiated. The list includes a probable new zeolite, related to phillipsite, for which the name 'garronite' is tentatively suggested.

TABLE I. Relative abundance of the amygdale minerals in the Tertiary lavas of Ireland.

Chabazite	84 % of localities
,, habit 1*	30
,, habit 2	52
,, habit 3	41
,, habit 4	19
,, habit 5	6
Thomsonite	53
Calcite †	about 40
Analcime	31
Levyne (and fibrous overgrowth)	27
Natrolite	23
Mesolite	10
Stilbite	9
Gmelinite	9
Heulandite	7
Phillipsite	6
Aragonite	6
Apophyllite, gyrolite	5
Quartz, chalcedony, opal	3
Gismondine	<1
'Garronite'	<1
Scolecite‡	<1
Laumontite	<1
Mordenite	<1
Localities with secondary minerals, but zeolites absent	5

* Following the chabazite habit notation of Walker (1951, p. 779).

† Calcite may originally have been much more widespread than this, much of it subsequently being dissolved away.

‡ Independent scolecite; scolecite as a thin coating on mesolite is more abundant.

The following descriptive data on the minerals are intended to supplement those given in the previous two papers of this series:

Thomsonite of faröelite habit is characteristic of the Antrim Basalts, and later bladed thomsonite of sphaerostilbite type is also widespread. Thomsonite of an unusual habit, best described as 'oolitic', occurs in two areas, one near Randalstown, and the other north of Templepatrick. The 'ooliths', pale brown in colour and individually 1 to 2 mm. in diameter, are compact spheres with radiating structure. They are grouped together either as a solid mass which may completely fill an amygdale, or as a loosely joined cluster partly or completely filling an amygdale, or sometimes forming a bedded floor to the cavity. This 'oolitic' thomsonite is optically quite normal. A similar habit of thomsonite, with pisolith-like bodies 5 mm. in diameter is developed occasionally in tholeiite lavas at Bengore Head. Pale brown in colour, the 'pisoliths' are loosely aggregated to form a bedded floor to large amygdales, with later colourless thomsonite (of faröelite habit) and chabazite.

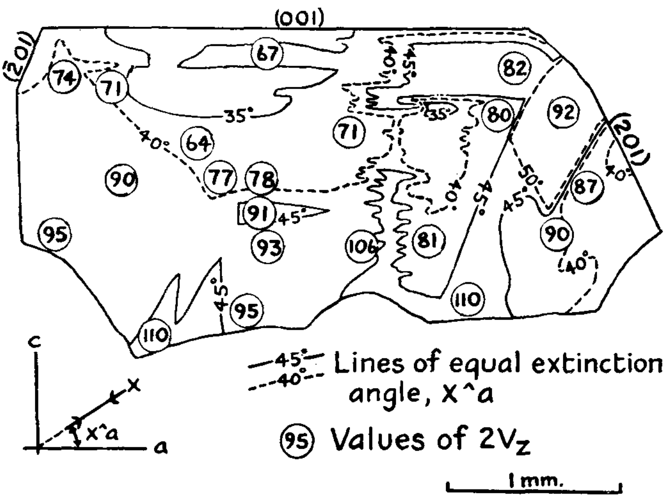

Fig. 1. Optical properties of a cleavage flake of heulandite from the Giant's Causeway area.

Stilbite forms both sheaf-like aggregates and single crystals platy parallel to {010}. All variations are found between crystals terminated by {110} and rectangular plates of epidesmine habit terminated by {$\bar{1}$01} alone. Unlike that in the Scottish Carboniferous basalts, the Antrim stilbite is invariably white or colourless. Optically it is monoclinic, with an extinction angle of 1 to 5°, showing sector twinning. The range of refringence observed is α 1·492–1·500, γ 1·502–1·513.

Heulandite always occurs in well-shaped crystals with little tendency to form parallel or radiating aggregates. The forms habitually present are {010}, {001}, {201}, {$\bar{2}$01}, and {110} and the habit varies but little, crystals from tholeiite in north Antrim being more elongated along *a* than those in south Antrim, which are elongated along *c*. The largest crystals seen measure about 5 mm. in length, the average size being only slightly smaller than this. The Antrim heulandite is always white or colourless.

Optically the heulandite is biaxial with the acute bisectrix, γ, normal to the {010} cleavage. The refringence covers a small range ($\alpha = 1{\cdot}491$–$1{\cdot}501$; $\beta = 1{\cdot}493$–$1{\cdot}503$) but the range may be almost as great as this within the confines of a single crystal, both concentric and sector zoning being developed. For example, in one crystal, β for the {001} sector was 1·498; for the {$\bar{2}$01} sector 1·497; and for a thin outer rim to both sectors 1·503. The extinction angle and optic axial angle are similarly variable and in a single crystal each may cover a range of 30° or more. On a typical crystal from the Giant's Causeway, for instance (Fig. 1) $2V_\gamma$ covers the range 64 to 110°; on another from south Antrim 0 to 35°. Some of these variations may be related to variations in the water content, for slight heating is sufficient to change the extinction angle markedly.

Phillipsite forms well-shaped crystals, and the Causeway tholeiites afford the best British localities for this mineral, crystals that exceptionally attain a length of 1·5 cm. being found there. Phillipsite also forms radiating aggregates, and these are especially common in the gmelinite-bearing lavas of Islandmagee and adjacent parts of east Antrim, in which phillipsite is unusual in being red in colour. Optically the Antrim phillipsite is biaxial positive, with moderate 2V; β ranges from 1·491 to 1·499 in the specimens studied, the lower values being found in the gmelinite-bearing lavas; γ–α is of the order 0·004.

Apophyllite shows a considerable range of crystal habit, but almost the entire range may be represented at a single quarry (fig. 3). The most characteristic habit simulates cubic symmetry, crystals being bounded by {100} and {001} and having the length of the {100} faces approximately the same as the width. Small faces of {111} are usually also present. Some crystals are clear and colourless, but usually some degree of alteration is seen, and all gradations are seen to pseudomorphs in a soft, white, greasy material.

Optically the apophyllite is sometimes uniaxial, sometimes biaxial with 2V up to 35°; in some crystals both uniaxial and biaxial sectors are found, as in the example illustrated (fig. 2). Determinations of refractive index for specimens from ten localities give a range in ω of 1·532 to 1·543. The optical sign is positive when ω lies below 1·540, and usually negative when above. The birefringence falls with increasing refringence, as noted by Wenzel (1917), from 0·004 when $\omega = 1{\cdot}532$ to less than 0·001 when ω lies above 1·540. All samples examined with $\omega = 1{\cdot}540$ or more show spectacular abnormal blue or purple polarization colours between crossed nicols due to the strong dispersion.

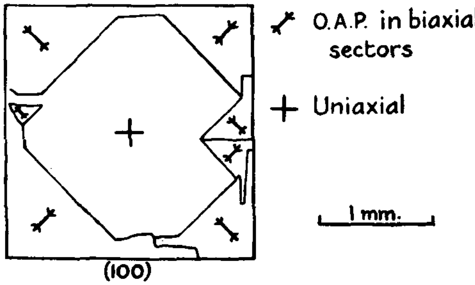

FIG. 2. Optical orientation of biaxial sectors about a uniaxial core in (001) cleavage plate of apophyllite from quarry on the north side of Carnmoney Hill.

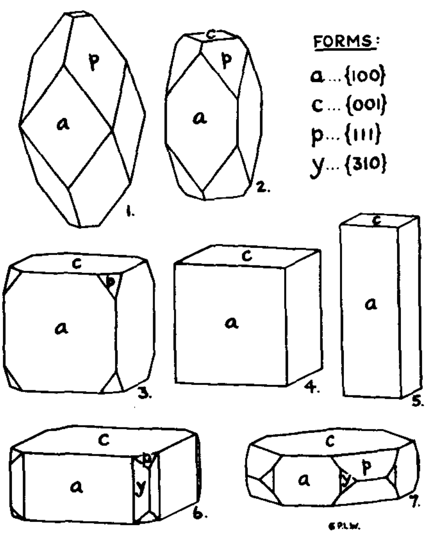

FIG. 3. Crystal habits of apophyllite encountered in a basalt quarry 1·1 mile north-east of Wolf Hill, Legoniel, Belfast. Habit 3 is most characteristic of Antrim apophyllite.

Zoned crystals normally show an increase in refringence and decrease in birefringence towards the margin, and sometimes the sign also changes. For example:

Berk Hill:	core	$\omega = 1{\cdot}536$,	$\epsilon - \omega = 0{\cdot}003$,	opt. + ;
	margin	$1{\cdot}543$,	$\omega - \epsilon < 0{\cdot}001$,	− ;
Roughfort:	core	$1{\cdot}537$,	$\epsilon - \omega = 0{\cdot}002$,	opt. + ;
	margin	$1{\cdot}542$,	$< 0{\cdot}001$,	+.

Gyrolite forms rosettes of mica-like flakes averaging 5 mm. in diameter, with pearly lustre parallel to the basal cleavage. Optically it is unaxial negative, and measured values of ω range usually from 1·544 to 1·554, although sometimes the refractive index is appreciably lower than 1·54.

Laumontite has been found by the writer at one locality in south Antrim as prismatic crystals 5 mm. in length bounded by {110} and $\{\bar{2}01\}$. It is in the partially dehydrated form leonhardite, and optically it is biaxial with $\alpha = 1{\cdot}508$, $\gamma =$ about 1·515, $\gamma{:}c\,\{001\}$ 42°.

Mordenite, although identified by the writer at only two localities,[1] may be more abundant than this. It occurs in aggregates of delicate fibres and has been identified by its low refractive index.

Mineral associations and zones.

The amygdale minerals are not distributed erratically through the lavas. They are readily grouped into five mineral assemblages, as listed below. The distribution of these assemblages can be, and has been, mapped in the field, and systematic mapping shows that the assemblages characterize definite zones. It should be pointed out at the outset, however, that the amygdale minerals in the tholeiite lavas of north Antrim do not fit into this scheme, and the present discussion applies only to the olivine-basalt lavas, which constitute the greater part of the Antrim lava pile.

The five mineral assemblages are as follows:

Stilbite–heulandite assemblage; essential minerals stilbite and heulandite. They may be associated with any of the other zeolites, gmelinite excepted. The most common associates are chabazite, calcite, analcime, and natrolite; thomsonite, apophyllite, and gyrolite also commonly occur.

Analcime–natrolite assemblage; essential minerals analcime or natrolite or both in the absence of stilbite, heulandite, and gmelinite. The most common associates are chabazite and calcite; apophyllite and gyrolite, and sometimes phillipsite and levyne may also be present.

Chabazite–thomsonite assemblage; characterized by the association of chabazite and thomsonite in the absence of gmelinite and of the index minerals of the two preceding assemblages. Levyne usually occurs as well, and associated minerals include phillipsite, calcite, and less commonly apophyllite and gyrolite.

Gmelinite assemblage; the essential feature is the presence of gmelinite,

[1] In a quarry ½ mile N.N.E. of summit of White Mountain; and in a shore exposure at Blackcave, Islandmagee

with which may be associated any of the other zeolites (in practice stilbite and heulandite are missing). The common associates are chabazite, thomsonite, analcime, phillipsite, levyne, calcite, and aragonite.

Plumose-calcite–aragonite–quartz assemblage; quartz, chalcedony, and opal are confined to this assemblage, and aragonite is much more abundant here than elsewhere. The calcite possesses a distinctive 'plumose' habit not found elsewhere, and $\{02\bar{2}1\}$ is the dominant form. Typically zeolites are completely absent, but there is a transitional assemblage in which zeolites are associated with an abundance of calcite, aragonite, and quartz.

The distribution of these five mineral assemblages is represented on the map, fig. 5. This map is based on data from 670 localities, and the degree of control achieved in locating the zone boundaries may be seen by comparison with the locality map, fig. 4. The chabazite–thomsonite assemblage is to be regarded as the most characteristic of the Antrim Basalt, for it covers the largest area and probably constitutes more than half of the volume of the lavas. It is followed downwards by the analcime–natrolite assemblage which, although covering a rather small area on the map, probably underlies the chabazite–thomsonite zone as a more or less continuous zone over most of the area of the basalts and may constitute nearly half of the total volume.

The distribution of the chabazite–thomsonite and analcime–natrolite assemblages.

The first clear picture of the distribution of the mineral assemblages was obtained by a detailed study of the well-exposed ground of the Garron Plateau and adjacent areas in eastern Antrim (Walker, 1951). Two assemblages are developed there, namely the analcime–natrolite assemblage, which almost everywhere forms a zone in the lower few hundred feet of the lava succession, and the chabazite–thomsonite assemblage, which everywhere overlies it and in a few places underlies it as well. Although conforming roughly to the stratification of the lavas, when examined in detail the zeolite zones are seen to be discordant and the same applies to the chabazite crystal habit zones that were mapped; this indicates clearly that the zeolites postdate the lavas in which they occur by a substantial margin.

It is clear that the general relations worked out in the Garron area apply to the greater part of the Antrim Basalts. An analcime–natrolite zone has been mapped around most of the periphery of the basalt plateau (fig. 5) and in places it is seen as small inliers within the plateau.

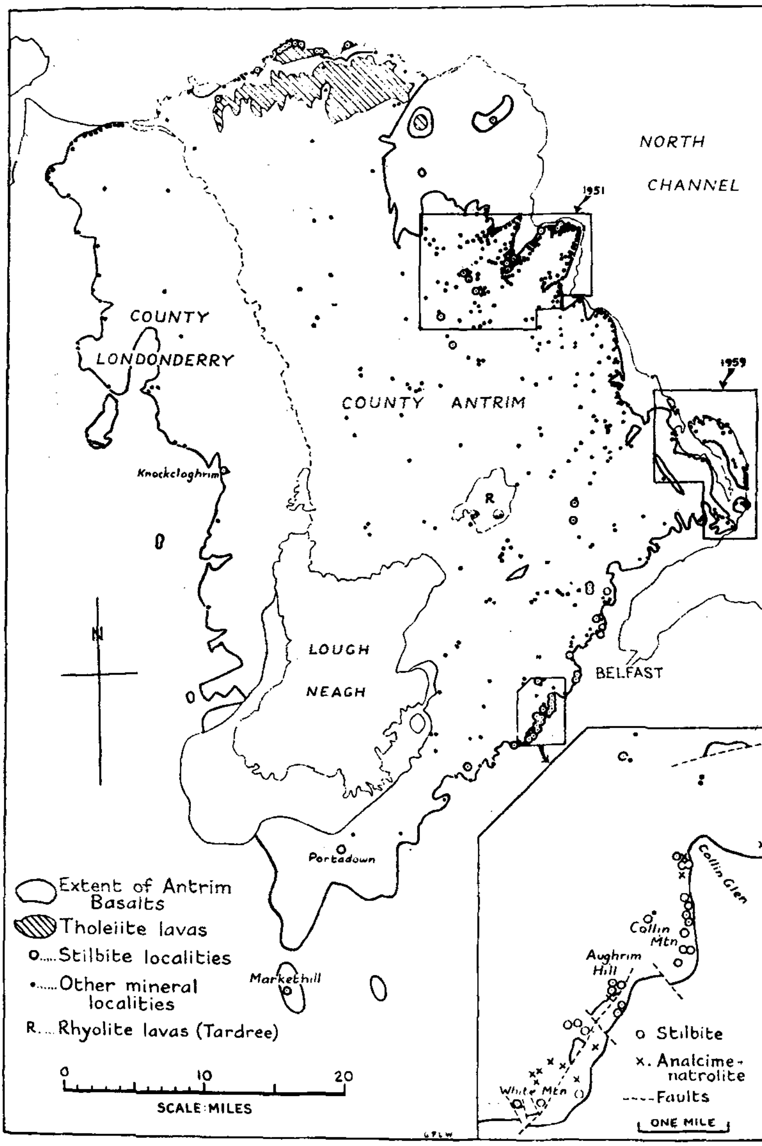

FIG. 4. Map of the Antrim Basalts showing the localities on which the zeolite distribution map is based, and also showing the distribution of stilbite. Detailed maps of the areas labelled '1951' and '1959' have been published in the two earlier papers of this series.

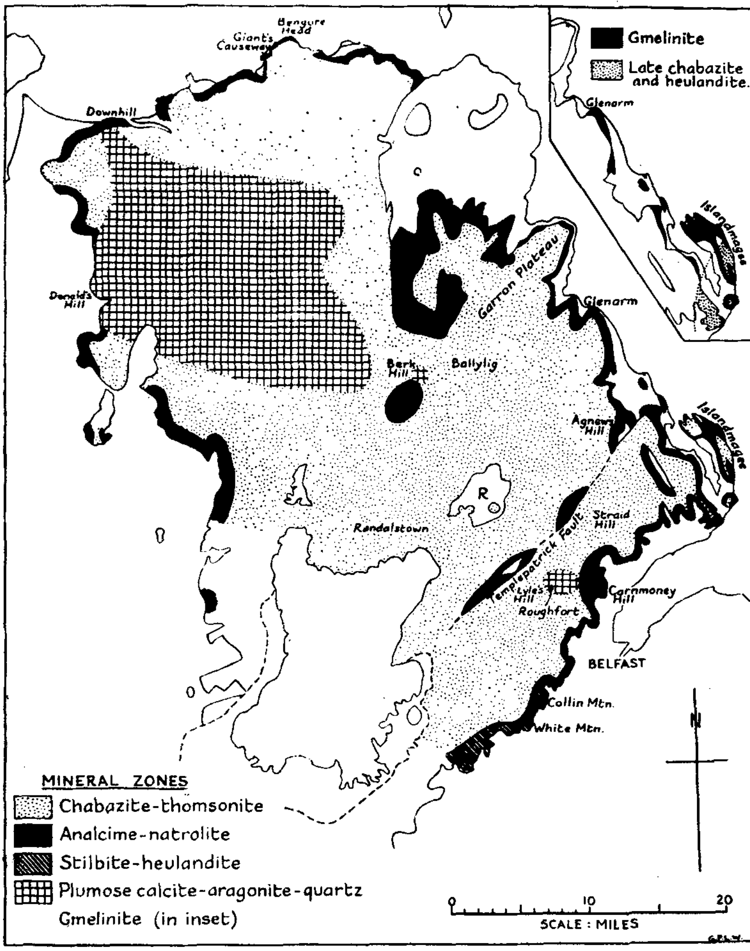

FIG. 5. Zeolite distribution map of the Antrim Basalts.

Inliers of the zone are also found along the northern, upthrow, side of the Templepatrick fault, where the lower parts of the basalt pile have been brought to the surface by faulting. Everywhere the zone is overlain by one with the chabazite–thomsonite assemblage.

Although mapped as part of the same zone, and clearly requiring closely similar conditions for their formation, there is a tendency for analcime and natrolite to be mutually exclusive or antipathetic to the extent that when one is abundant the other usually is not. Analcime is predominant along the north coast from Downhill to the Causeway (fig. 6); along the north and east of the Garron Plateau to as far south as Carnlough; and in south Antrim south-westwards from Belfast.

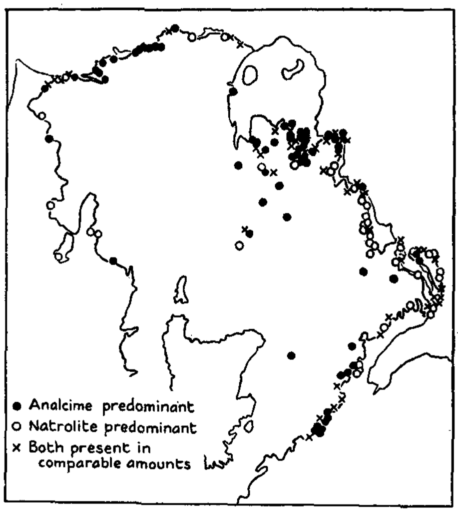

FIG. 6. The distribution of analcime and natrolite in the analcime–natrolite zone.

Natrolite is predominant in eastern Antrim at most localities from Carnlough to Belfast; along the north coast near Whitepark Bay and at Downhill; and in the west near Carntogher.

The distribution of chabazite crystal habits likewise shows everywhere a general pattern similar to that worked out for the Garron area, although exposures are seldom sufficiently numerous to enable a habit distribution map to be drawn. In the Garron Plateau and elsewhere those localities bearing chabazite crystals of more complex habit (habits 4 and 5, Walker, 1951) have a distribution very similar to that of the analcime–natrolite zone, being found in the lower few hundred feet of the lavas and distributed around the periphery of the basalt outcrop; these complex habits are also seen in a few inliers, such as that of Berk Hill and the ground to the north. The crystal habit of chabazite changes progressively upwards and crystals of the simplest habits (1 and 2) are characteristic of the higher parts of the basalt pile, for example, Agnew's Hill and the higher parts of the Garron Plateau.

Herschelite (chabazite of phacolite habit with {0001} developed) has been found at a single locality low down in the basalts, a path exposure in a slip on the basalt escarpment ½ mile south of Drumnagreagh House, south-east of Glenarm. The herschelite is associated with chabazite of lower habits (3 or 4) and a little later chabazite of habit 2, together with natrolite.

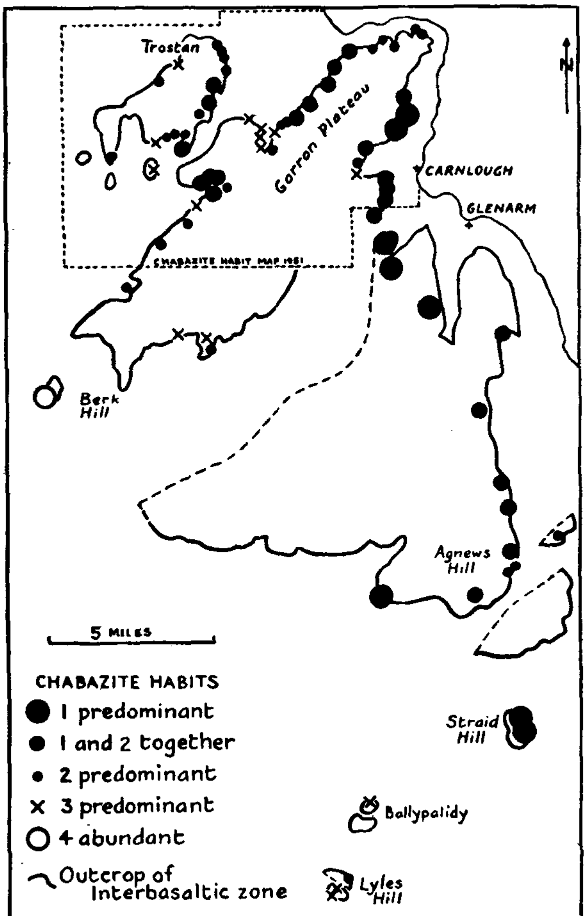

FIG. 7. The crystal habit of chabazite in lavas at the base of the Upper Basalts, eastern Antrim.

In eastern Antrim there is a tendency for the chabazite 'isohabit surfaces'[1] to rise westwards or south-westwards through the lavas.

[1] The term 'isohabit' was suggested in the 1951 paper for lines of equal crystal habit as drawn on a map of the Garron Plateau area.

Thus, if one takes a recognizable stratigraphic horizon, the base of the Upper Basalts (fig. 7), habits 1 and 2 are the only habits found at this level in the east of the region (the eastern part of the Garron Plateau; the areas near Carnlough; Glenarm; Scawt Hill; Agnew's Hill; and Straid Hill), while habits 4 and 5 are found in the west (Trostan; Ballylig; Berk Hill; Ballypalidy; and Lyle's Hill). The higher habits are not found at all in the basalts farther east.

The distribution of stilbite and heulandite.

Where the analcime–natrolite zone near the base of the lavas is thickest, these two minerals are joined by stilbite and heulandite; where these latter minerals are abundant there is justification for the recognition of a separate mineral assemblage, the stilbite–heulandite assemblage. This assemblage is well developed only in the south of Antrim, along the escarpment south-westwards from Belfast, and is best seen in that stretch from Collin Glen to the White Mountain (figs. 4 and 5). There are a few exposures farther towards the south-west (Maghaberry, Portadown, and Markethill) that also contain stilbite or heulandite, but exposures are too scattered to be certain that the stilbite–heulandite zone extends continuously in that direction.

In the stretch from Collin Glen to White Mountain, stilbite and heulandite are extremely abundant in a number of quarries in the lower 400 feet of lavas, and they are accompanied by chabazite, often of habits 3 and 4, analcime, natrolite, and by much calcite, often brown. These minerals are usually found in the red weathered amygdaloidal top and base of a lava flow, while other minerals, namely thomsonite, chabazite, apophyllite, and gyrolite are met with in the less abundant amygdales in the fresh interior of the flow (see p. 522). Thomsonite, although common, is almost never found associated with stilbite or heulandite in the same amygdale; these minerals are segregated in different parts of the same lava flow. Likewise it is extremely rare for apophyllite or gyrolite to appear in the same cavity as stilbite or heulandite.

The age relations of the minerals are in some doubt. Heulandite, when associated with stilbite, is usually found as small, well-shaped crystals around the walls of the cavity, embedded in stilbite, which mineral is thus probably later. The most common associate of stilbite is chabazite of habit 2 to 3. Sometimes the chabazite crystals have been deposited on the stilbite, and sometimes the reverse is true, but more often there is no reliable indication of the relative age and it seems as though the two minerals have crystallized more or less simultaneously, or their periods of crystallization have overlapped. Analcime is invariably later than stilbite.

While exposures in the stilbite–heulandite zone of the area around White Mountain and Collin Glen are generally excellent, they fail at critical points, and the shape of the zone is not certain. Exposures are not sufficiently good to establish that the stilbite–heulandite zone is flat-lying and does not cut steeply across the lava stratification, and the main evidence favouring the former arrangement is the restriction of stilbite and heulandite to a very narrow belt, nowhere more than a mile in width, along the basalt escarpment where the lower parts of the lava succession are exposed.

If one assumes, then, that the zone is flat-lying, it must attain its greatest known thickness, around 400 feet, on Collin Mountain. On the White Mountain the thickness has probably decreased to around 100 feet. There is an indication that the stilbite–heulandite assemblage is underlain and overlain by a narrow zone characterized by analcime and natrolite and that the former zone forms in fact the core of the analcime–natrolite zone. Traced north-eastwards towards Belfast it passes into the typical natrolite–analcime assemblage, with sporadic stilbite or heulandite in small amount.

The great bulk of the stilbite and heulandite in Antrim is in the region of White Mountain and Collin Glen, and there is no comparable zone elsewhere. Stilbite occurs sparsely at a number of localities on the edge of, or near, the Garron Plateau (fig. 4), most of them in the Lower Basalts near their base (e.g. in Glenariff Glen) within the analcime–natrolite zone. Stilbite is abundant at a large quarry at Knockcloughrim at the western edge of the basalt outcrop; heulandite is sparsely developed at Portballintrae on the north coast; and stilbite is seen on the upthrow side of the Templepatrick fault, near the base of the lavas at Ballyclare.

At one locality in the stilbite–heulandite zone, a quarry $\frac{1}{4}$ mile north-west of Castle Robin and $\frac{3}{4}$ mile north-east of the White Mountain, crystal cavities several centimetres in length were found in large amygdales with later analcime growing in the cavities. The shape, although unlike that of calcite, is not sufficiently well-defined to enable positive identification of the mineral leached out. The general characters remind one of the cavities due to anhydrite in the Watchung basalts (Schaller, 1932). Laumontite (leonhardite) has been observed at one locality in the stilbite–heulandite zone (Belshaw's Quarry, one mile south-west of the White Mountain) in small amount in the lowest lava flow, associated with chabazite, stilbite, and heulandite.

The distribution of apophyllite and gyrolite.

Apophyllite and gyrolite, a pair of minerals seldom seen apart, have been found at some thirty localities (fig. 8). Most are in the south-east of Antrim, especially near Belfast and in the White Mountain area, and

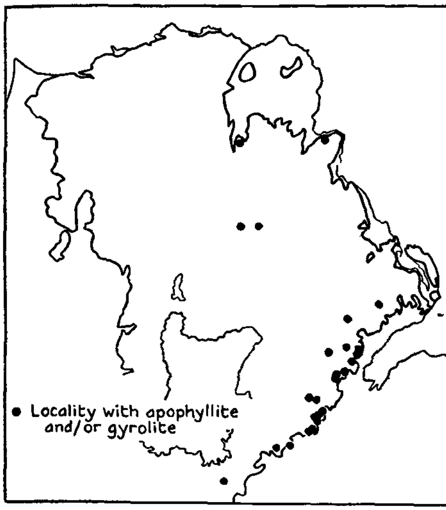

FIG. 8. The distribution of apophyllite and gyrolite in the Antrim Basalts.

the most important are: *Belfast area*: Quarries on Catcairn and Wolf Hill; on the northern and western side of Carnmoney Hill (one described by Fleischmann, 1910); abandoned quarries (Fleischmann, 1910) at Ballyhenry; *White Mountain area*: abundant gyrolite in quarries near Castlerobin; sparse apophyllite in quarry north-west of summit of Collin Mountain; *Other areas*: Quarries in Upper Basalts, Berk Hill.

Very fine collections of apophyllite from quarries near Belfast were made by the late Robert Bell and F. N. Ashcroft in the first decades of this century. They include the best British examples of the mineral, the crystals commonly exceeding 1 cm. in size. Apophyllite and gyrolite are virtually confined to the often very large and horizontally flattened cavities in the black interior of massive basalt lava flows, and as such cavities are seldom numerous and the fresh basalt is very tough good specimens can in practice be obtained only in quarries that are actively worked.

Specimens from these localities have a markedly uniform appearance. The cavities have a compact radiating-fibrous lining of faröelite, and apophyllite forms well-shaped crystals, sometimes water-clear, at other times dull white, resting on it. Closely associated, and also later than the faröelite, are rosettes of pearly-white leaves of gyrolite, and little sheaves and tufts of thomsonite of sphaerostilbite habit or, less commonly, thomsonite in radiating clusters of separate needles or fibres. Sometimes there are tiny brown crystals of calcite as well. Adjacent cavities may contain faröelite, levyne, or chabazite.

Apophyllite is invariably later than thomsonite of faröelite habit; the age relationship to the other zeolites is seldom apparent but several examples have been found of apophyllite following chabazite and stilbite, and one with small crystals of apophyllite encrusting analcime.

Apophyllite and gyrolite have been found in the stilbite–heulandite, analcime–natrolite, and chabazite–thomsonite assemblages, most of the localities being in the south-east of Antrim. Before considering the possible zonal distribution, however, one must take account of the following significant observations:

With two exceptions, apophyllite and gyrolite have been encountered only in quarries; they are practically confined to the massive black interior of lava flows, being very rare in the red amygdaloidal tops; and apophyllite is manifestly one of the least stable of the minerals, the water-clear crystals readily becoming white and chalky on weathering (cf. Bailey, 1941).

These observations are interpreted as meaning that apophyllite is readily destroyed by weathering and the known distribution may bear little or no relationship to the actual distribution just below the zone of surface weathering; apophyllite has been destroyed in practically all natural exposures and is seen only where quarries reach below the surface zone of weathering, as in the quarries that are so numerous in south-east Antrim. Even in these quarries apophyllite may have been destroyed in the more permeable red amygdaloidal tops of the lava flows, for it is now seen only in the massive interior of the flows.

The distribution of gmelinite.

Gmelinite is restricted to a narrow zone along the eastern seaboard of Antrim in which it is extremely abundant. Its distribution has already been described and discussed (Walker, 1959*a*).

The distribution of the calcite–aragonite–quartz assemblage.

The variety of calcite referred to in this paper as 'plumose' forms more or less radiating aggregates of crystals with curved cleavage surfaces, and the crystals have grown in the direction of one of the Miller crystallographic axes, with two cleavages disposed approximately normal to the rock-surface from which the calcite grew. Plumose calcite occurs over a large and well-defined area in the north-west of the basalt outcrop as an important mineral in a well-defined mineral assemblage. In it the calcite is characteristically associated with aragonite and commonly with quartz, chalcedony, opal, and macroscopic chlorite; usually calcite of normal habit and sometimes pisolitic calcite and 'satin spar' (fibrous calcite) are also present. Zeolites are completely absent.

The plumose calcite is often pale green in colour, and usually cloudy or milky. Curved crystal faces are often seen terminating crystals in the aggregates; these faces are seldom bright, but it is clear that $\{02\bar{2}1\}$ is the predominant and often the only form developed. Quite commonly normal calcite, clear and colourless and with plane cleavage surfaces, continues the growth of the plumose calcite as a parallel overgrowth.

Aragonite typically forms solid radiating columnar aggregates filling joints and amygdales, and exceptionally the individuals in these aggregates attain a length of over one foot. Aragonite in these aggregates is often pale pink or lilac in colour. Frequently it is largely or entirely replaced by a sugar-grained aggregate of calcite, the radiating form of the original mineral being preserved in the paramorphs.

Very rarely delicate acicular crystals of aragonite are encountered as, for instance, in a basalt quarry 2 miles SSE. of Donald's Hill. The basalt here contains cavities up to a foot or more in diameter, which are lined with a layer half an inch thick of green plumose calcite, superficially very like prehnite. This is succeeded by a layer of chalcedony, and that in turn by quartz, and the slender prisms of aragonite occur in cavities in the quartz, accompanied by a little normal calcite.

Quartz and chalcedony, apart from their occurrence in tholeiites in north Antrim, are practically confined in the Antrim Basalts to the plumose-calcite zone. In this zone they are both abundant, and are sometimes accompanied by a little milky or hyaline opal. These minerals are later than the bulk of the calcite.

This assemblage of minerals is present over a large area of lavas in the north of County Londonderry and adjacent parts of Antrim. The boundaries of this zone are fairly reliably localized along the western escarpment in Co. Londonderry, but elsewhere exposures are not sufficiently abundant to enable this to be done; no exposures have, for example, been found in the broad and low-lying valley of the river Bann, which crosses the outcrop of the zone. Around the margins of the plumose-calcite–aragonite–quartz zone, from which zeolites are absent, a transitional zone is found in which the minerals are accompanied by zeolites. The arm of the zone shown on the map, fig. 5, extending towards the north coast near Downhill is inferred from the abundance of carbonates in the zeolite-bearing assemblages on the coast there.

An assemblage of minerals similar to that of the plumose-calcite zone is seen at a single quarry north of Ballymena, just east of the Berk Hill outlier, and at a number of quarries near the hamlet of Roughfort, east of Templepatrick, in south Antrim.

The three-dimensional shape of the plumose-calcite zone is not known, mainly on account of the shortage of suitable exposures. The characteristic mineral assemblage of the zone is certainly developed through a vertical thickness of up to 500 feet of lavas, and nowhere has evidence been found of its being underlain or overlain by zeolite-bearing zones. The origin is likewise uncertain. The arguments that show that formation of zeolites took place long after the exposed lavas had been erupted apply with equal force here. One can envisage the minerals being formed at a late stage in the volcanic history of the area by regional thermal waters perhaps rising from some deep-seated source but there is no indication of what that source might be.

In the Tertiary lava region of eastern Iceland a very similar mineral assemblage is encountered in the vicinity of major volcanic centres; there the calcite, aragonite, quartz, and other minerals, are clearly due to heated solutions or gases coming from those centres and permeating the adjacent lavas. It is tempting to see in the plumose-calcite–aragonite–quartz zone of the Antrim Basalts evidence of a comparable Tertiary volcanic centre. However, the available geological evidence is opposed to such a possibility (p. 525).

Minerals in the Causeway tholeiites.

In the Antrim Basalts tholeiite lavas are confined to the coastal strip along the north of Antrim, where there is a group of thick flows with a maximum aggregate thickness of about 450 feet (Tomkeieff, 1940; Patterson, 1955). The group, referred to in the older literature as the 'Upper Basalts', was renamed the Middle Basalts by Patterson who showed that they were underlain and overlain by olivine-basalts (the Lower and Upper Basalts of Patterson), the former ranging in thickness from 300 feet near Ballycastle and Portrush to less than 100 feet near Carrickarade. The uppermost part of the Lower Basalts had suffered deep lateritic weathering prior to the eruption of the tholeiite flows.

The tholeiite lavas are famous for their columnar jointing, and are well exposed in the coastal cliffs from the Giant's Causeway to Bengore Head. These cliffs, up to 360 feet high, are accessible by cliff-path, and the screes from them contain very fine specimens of the amygdale-minerals; other localities from which mineral specimens may be obtained from the tholeiites include Craigahulliar quarry, near Portrush, and, according to the literature, Rathlin Island. The Giant's Causeway is often given as a mineral locality in the literature although, as Ashcroft (1916) has pointed out, the term 'Giant's Causeway' was often used in the widest sense to indicate almost any locality along the north coast of Antrim.

The following is a list of the minerals recorded in the literature, probably from tholeiite lavas, in the Causeway area: analcime, apophyllite, brewsterite, chabazite, harmotome, heulandite, mesolite, phillipsite, scolecite, stilbite, and thomsonite.

The writer has not found apophyllite, brewsterite, or harmotome, and suspects that chabazite was mis-identified as apophyllite, phillipsite as harmotome (these two minerals being indistinguishable in the hand-specimen), and stilbite as brewsterite; stilbite resembles brewsterite very closely when terminated by $\{10\bar{1}\}$ alone. To the list can now be added levyne, which although rare does undoubtedly occur, together with the fibrous overgrowth that is so characteristic of much of the Antrim levyne; also natrolite, opal, and celadonite; and possibly laumontite.

The tholeiite lavas, like the olivine-basalts, generally have amygdales in the upper half or third of the flow, culminating in a highly amygdaloidal red topmost part, but the amygdale-minerals exhibit certain differences when compared with those in the olivine-basalts. The first difference is the presence of celadonite, a dark green mineraloid that is confined to the tholeiites. It forms amygdale-linings in the interior of a flow below its red top. When this lining weathers, the amygdales separate from the basalt and resemble almonds much more closely than they ever do in the olivine-basalts, where amygdale and basalt are firmly knit together. Occasional cavities in the tholeiites are completely filled with celadonite.

A second distinctive character is the abundance of quartz, chalcedony, and opal in the tholeiites. Outside the plumose-calcite zone, these silica minerals are almost never encountered in the olivine-basalts. Thirdly, stilbite and heulandite have a rather different habit in the tholeiite lavas. Stilbite is in small crystals with square cross-section or in hemispherical aggregates with smooth outer surface, and is usually terminated by $\{10\bar{1}\}$ alone; in the olivine-basalts it is in large, platy crystals or in large sheaf-like aggregates more commonly terminated by $\{110\}$. Heulandite in the tholeiites is generally more elongated parallel to *a* than in the olivine-basalts.

A marked difference in the assemblage of amygdale minerals is found in the two types of basalt where they are contiguous, as in the Causeway cliffs. The olivine-basalts bear chabazite, analcime, natrolite, and calcite, with only traces of other minerals. The overlying tholeiites contain in addition to these minerals an abundance of stilbite, heulandite, and phillipsite at most localities, as well as celadonite and silica minerals. Indeed, one can quite reliably map the two basalt types solely on the basis of their secondary minerals. This is the only known instance in the Antrim Basalts where the composition of the lava obviously exercises a control on the mineral assemblage formed. A similar relationship is known in other areas, for instance in Iceland and Mozambique (Holmes, 1916).

A form of zonal arrangement of amygdale-minerals within the confines of a single lava flow is often observed in olivine-basalts with, for instance, thomsonite confined to the fresh interior of a flow and stilbite and heulandite to the red amygdaloidal top (p. 515). Such zoning reflects the different permeability of the massive interior of a flow and the slaggy top. In the tholeiite flows a zonal arrangement is nearly always present, with chalcedony and quartz confined to the black interior and zeolites confined to the more highly vesicular upper parts.

Occasionally a much more striking zoning is seen in the tholeiites. Careful examination of fallen blocks on the western side of Benanouran Head reveals the following zonal arrangement in a single lava flow:

Red decomposed amygdaloidal top: much calcite, chabazite.

Rather decomposed amygdaloidal basalt below red top: chabazite and phillipsite.

Interior of flow (upper part), with few cavities, horizontally flattened by flow: celadonite, stilbite, heulandite, chabazite, and phillipsite. Chabazite and phillipsite are both probably absent from the lower part of this zone.

Interior of flow, sparse cavities, horizontally flattened by flow: celadonite, chalcedony, quartz, opal; zeolites absent.

Lower half of flow, free from cavities: amygdale-minerals absent.

It was at first thought that this zonal arrangement might be due to mineralization during the cooling of the flow on the surface, but this idea was abandoned in face of the overwhelming evidence that in general zeolitization is later. It is more probably related to the progressive upward increase in porosity and permeability of the basalt.

Zeolites in dolerite intrusions.

Numerous dolerite intrusions—dykes, plugs, and sills—are found associated with the lavas. Most are of olivine-dolerite or crinanite, similar in composition to the lavas, and of the same general age. The dolerites usually contain zeolites, often forming around 5 per cent. of the volume of the rock. The zeolites occupy interstitial patches between the pyrogenetic minerals, and to a certain extent they replace these minerals; in some dolerites zeolites are also found filling small amygdales in the rock. Dolerite pegmatites are generally richer in zeolites than the main dolerite body in which they occur.

The zeolites in the several dolerite intrusions that have been studied by the writer are similar to those in the adjacent lavas, which comprise the country rock. Those intrusions in the lower parts of the lava pile and in the subjacent sedimentary rocks contain an assemblage of zeolites that includes prominent analcime or natrolite. Intrusions high in the lava pile lack analcime and natrolite and bear the minerals typical of the chabazite–thomsonite assemblage. Thus the Parkgate plug (Walker, 1959*b*) contains an abundance of levyne, together with chabazite and phillipsite, and the dolerite is locally veined by thomsonite and chabazite; the Ticloy plug contains phillipsite, chabazite, rare levyne, and a little probable analcime; and the Donegore 'dyke' bears levyne and thomsonite. All three plugs cut lavas in which the thomsonite–chabazite assemblage is developed. The conclusion appears inescapable that zeolites in dolerites and in the country rock were formed at one and the same time.

Discussion.

Perhaps the main result of this work has been to show that the distribution of amygdale-minerals is orderly and capable of being mapped; that the minerals occur in zones that are mostly flat-lying and nearly parallel to the stratification of the lavas, but in detail are seen to be discordant. All the evidence indicates that zeolitization took place at a time probably long subsequent to the eruption of the individual lava flows in which the minerals occur, and after at least 1 000 feet of lavas had been erupted, and locally it is possible to relate the period of zeolitization to the period of main movement along faults (Walker, 1951 and 1959*a*). There seems no direct means at present of determining in Antrim how much of the lava pile has subsequently been eroded away, but in Iceland, where the zeolite zones are broadly similar to those in Antrim, there is evidence that the original top of the lava pile stood some 2 000 feet above the top of the analcime zone. If this were so in Antrim it would indicate an original thickness of about 2 500 feet for the lavas in most of eastern Antrim, as compared with the present maximum exposed thickness of about 1 000 feet.

It seems possible to group the secondary mineral zones of the olivine-basalts into two categories: the normal zones, which are regional in extent and which are nearly parallel to the lava stratification; and the local zones, which are markedly discordant and of limited distribution. Into the first category come the zones of the Garron Plateau area and their extension elsewhere; into the second come the gmelinite zone and the plumose-calcite–aragonite–quartz zone.

The normal zoning consists of a chabazite–thomsonite assemblage underlain by one in which chabazite and thomsonite are joined by analcime and natrolite. In some places these minerals in the lower zone are joined by stilbite and heulandite, and where sufficiently abundant a distinct stilbite–heulandite zone may be distinguished. Broadly, the intensity of zeolitization increases as the number of mineral species present increases, and it is believed that these zones reflect a temperature control, the most intense zeolitization and highest temperature being attained in the stilbite–heulandite zone. It is possible that the flat-lying nature of the zones may be due to their being roughly parallel to the original top of the lava pile at the time when zeolitization was taking place; in any case it is to be expected in a pile of lava flows that are characteristically extremely permeable laterally along the slaggy tops of the flows but relatively impermeable in a direction normal to the stratification.

The writer considers that zeolitization got under way only after a certain critical thickness of lavas had accumulated, the blanketing effect of the upper lavas permitting the required build-up of heat in the lower flows. Antrim supplies no data on the value of this critical thickness, but Icelandic experience suggests that it may be of the order of 500 to 1 000 feet. Zeolitization took place in waterlogged basalt in which the water was of meteoric origin. The heat required came partly from below, and partly from exothermic hydration reactions, including the hydration of olivine, in the basalts; substantial increments also came from dykes and other bodies intruded into the lavas. Some of the faults cutting the

Garron Plateau (Walker, 1951, p. 788) apparently allowed heat to escape, and adjacent to them the higher-temperature analcime–natrolite assemblage is missing owing to this loss of heat.

The abnormal zoning is more difficult of explanation, and one must search for geological clues to account for it. The gmelinite zone has been shown to approximate in its position to a major fault zone, and it is reasonable to suspect a genetic connexion between the two, although it is difficult to account for the absence of gmelinite in close proximity to many another fault cutting lavas in Antrim and elsewhere. The agency of heated waters rising from depth may be responsible, but there are other possibilities. One is that the lavas containing gmelinite may have suffered an incursion of sea water during zeolitization, in place of the more usual meteoric waters. Geological evidence does no more than leave open the possibility that incursion of sea water may have been consequent on the downwarping of the lavas in the Islandmagee fault zone before or during faulting. It is for future experimental work to show whether the presence of chlorides in solution promotes crystallization of gmelinite or not, and whether sea water is responsible for the distinctive pink or red coloration of the gmelinite and other minerals in the gmelinite zone.

The abnormal plumose-calcite–aragonite–quartz zone is of the type that might be expected to develop about a locus of hydrothermal activity such as a volcanic centre. Certainly an assemblage very similar in character is encountered in the Tertiary basalts of eastern Iceland about major volcanic centres. However there is a complete absence of independent geological evidence for such a centre in the Antrim Basalts; the intensity of the dyke swarm, usually a sensitive indicator of such centres, is, indeed, lower here than almost anywhere else in Antrim (cf. map in Walker, 1959*b*; Patterson, 1955). The only geological abnormality known to the writer is the presence of an unusually well-developed set of north-west-trending joints in parts of the plumose-calcite zone. It is suggested that the minerals of this zone may have formed under conditions of free circulation of water facilitated by the joints in place of the more stagnant conditions favouring zeolites.

The observation that dolerite or crinanite intrusions (plugs, sills, and dykes) in Antrim bear the same zeolites as the lavas they cut is of considerable significance in connexion with the origin of zeolites in crinanites. It has usually been assumed that the analcime (and presumably the other zeolites that usually occur in crinanites as well) is a primary mineral or is formed during the cooling of the rock. In the Antrim intrusions on the other hand, it seems clear that, as in the lavas, the zeolites are entirely later than the rock in which they occur. The basalt lavas or the dolerite intrusions merely supply the cavities in which zeolites subsequently crystallize, plus the required chemical raw materials.

Finally, the mapping of the crystal habit of chabazite presents some of the most cogent geological evidence yet found to show that the crystal habit of a mineral, and the twinning, are not due to chance but are controlled by the environment under which the mineral crystallized.

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Appendix. The Langford Lodge borehole (Fowler, 1957) passed through some 2 590 ft. of basalts, the greatest thickness known in Antrim. The writer is indebted to the Director of the Geological Survey of Northern Ireland for permission to examine specimens from the borehole, and to publish this note. This part of the borehole was not cored, and the interpretation of the mineral distribution is open to doubt, but a fairly well-defined zonal distribution may be detected.

Chabazite is distributed throughout the borehole and is probably the most abundant zeolite present. In all but the uppermost 300 ft., in which it occurs sporadically, thomsonite is likewise distributed throughout. Levyne is present in most of the samples down to a depth of 1 300 ft., and little is found below that depth. Phillipsite occurs sporadically down to 1 000 ft. Stilbite is first encountered at a depth of 600 ft., and is abundant below 1 000 ft. Mesolite (with scolecite) is sporadically developed below 1 000 ft., but becomes very prominent below 1 700 ft.

In some respects the distribution resembles that found elsewhere in Antrim. However, stilbite is found through a much greater thickness than elsewhere (it is abundant in the lower 1 500 ft. in the borehole), and mesolite, never prominent in surface exposures, is very abundant in the lower 800 ft. in the borehole. Laumontite, extremely rare in surface exposures, is found at several levels below 1 800 ft. These differences may be accounted for by the supposition that the lavas in the Langford Lodge area not only attain an unusually great thickness at present, but originally had a much greater thickness there than elsewhere; the zone containing stilbite, for instance, is consequently 4 or 5 times as thick as it is in the Collin Mtn. area. Indeed, the observed distribution in the borehole bears a much closer resemblance to that in the Tertiary basalts of eastern Iceland, where a much greater thickness of lavas is exposed, and where stilbite and mesolite are both distributed through more than 2 000 ft. of lavas.

It is interesting to note that saponite and calcite are abundant throughout the borehole. This indicates clearly that they cannot be the result of present-day weathering of the lavas.