# PETROLOGY OF "EQUILIBRATED" CHONDRITES 2. METAMORPHISM AND THERMAL HISTORY

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*Abstract*: Petrological studies based mainly on the microprobe analyses have been made on nine ordinary chondrites, Yamato-74115 (H5), -74155 (H4), ALHA77148 (H6), Yamato-74007 (L6), -74190 (L6), -74354 (L6), -74362 (L6), -74445 (L6), and -74646 (LL6) and preliminary studies on the "unequilibrated" chondrites, ALHA77015 (L) and 77033 (LL).

Detailed microprobe analyses of minerals of chondrites indicate that some chondrules became rich in FeO from the surface after their formation and before accretion to form a parental body or bodies. Based on the textural and petrochemical data, thermal history of "equilibrated" chondrites has been inferred. It is suggested that highly recrystallized chondrites were derived from the relatively inner part of the parental body and weakly recrystallized ones from near the surface.

# I. Introduction

"Equilibrated" chondrites are believed to have been derived from "unequilibrated" chondrites by thermal metamorphism. Two types of metamorphism have been considered; (1) reheating of an initially cold body or bodies perhaps by decay of radioisotopes, the process called "prograde" metamorphism (VAN SCHMUS and WOOD, 1967; WOOD, 1967; DOOD, 1969), and (2) cooling of an initially hot body or bodies after the accretion, the process called "autometamorphism" or "retrograde" metamorphism (WASSON, 1974; HEYSE, 1978; ASHWORTH, 1980). It is still questionable, however, whether the "equilibrated" chondrites were derived from the "unequilibrated" chondrites. It is also not well understood what the "petrologic type" by VAN SCHMUS and WOOD (1967) means. Furthermore, their classification contains a problem that the petrologic type for one chondrite can not be defined uniquely.

The difference between "type 3" and "type 4" or between "unequilibrated" and "equilibrated" chondrites is important, because this difference corresponds to whether the process of homogenization of silicate minerals had occurred or not. For understanding the genetical relations between them, especially for the meaning of the "petrologic type", three H, five L and one LL ordinary chondrites with two unequilibrated chondrites have been studied in the present study. Based on their

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texture, microprobe analyses of silicate minerals and metals, and the equilibrium conditions, the thermal history of these chondrites are discussed. Petrographic studies of Yamato-74115, -74190, -74354, -74362, -74646 have already been done (NAGAHARA, 1979a) and brief descriptions of Yamato-74007, -74155, -74445 and ALHA77148, 77015, 77033 are given in Appendix I in this paper.

# 2. Iron Enrichment and Accretion of Chondrules

# 2.1. Oxidation of chondrules

After formation of chondrules and before accretion to a parental body, some chondrules are thought to become rich in FeO from the surface. Fig. 1 shows the zonal pattern of Mg and Fe in olivine grains in ALHA77015 (L3) chondrite. The grain in contact with the matrix shows an asymmetrical pattern; the rim which is in direct contact with the matrix is remarkably enriched in FeO, whereas the opposite rim is much less. On the other hand, the grain within a chondrule has a symmetrical zonal pattern and its FeO content is much less than the former grain.



The chemical composition of olivine cores in several porphyritic chondrules in ALHA77015 chondrite whose phenocrysts are olivine is shown in Fig. 2. Some of these olivine grains are within chondrules and the others are in contact with the matrix. The grains in contact with the matrix are richer in FeO than those within the interior of the chondrules. The composition of olivine is variable from grain to grain both within a single chondrule and among different chondrules; however, the average composition of olivine grains in the interior of a chondrule is obviously more enriched in MgO than that of grains at the rim.

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Fig. 2. Composition of core of olivine grains in some porphyritic chondrules in ALHA77015 chondrite. One diagram shows one chondrule. Square denotes olivine contacting with the matrix, and square with cross denotes that within a chondrule.

From these observations, it is considered that FeO or oxidized metallic iron on the surface of the chondrules diffused into them and olivine in contact with the matrix became iron-rich. As mentioned above, the composition of olivine is different among the chondrules, indicating that the original composition and the state of oxidation were different among different chondrules, and some of the chondrules may have become enriched in FeO.

# 2.2. Constraints for the accretion process

KIMURA et al. (1979) have already pointed out that the iron-rich rim had been attached to the chondrules before the accretion. Though the processes of the formation and accretion of the chondrules have not yet been clear, the above observations suggest that there might have been the conditions where the iron-rich rim of chondrules was oxidized.

Iron enrichment of the chondrules should have occurred before the accretion to form a parental body or bodies. Fig. 3 shows the chemical composition of the chondrites based on their chemical group and petrologic type by VAN SCHMUS and WOOD (1967). The figure indicates that the chemical composition of the chondrites, especially the contents of FeO,  $Fe^{\circ}$  (metallic iron) and FeS, which belong to the same chemical group, is nearly isochemical through the petrologic type. The state of oxidation, therefore, would not have changed after the accretion, so that the oxidation should have occurred before the accretion, perhaps in the nebula.

According to the experiments on the conditions of chondrule formation (TSUCHI-



Fig. 3. Elemental abundances of chondrites of each chemical group against the petrologic type. The numbers on the abscissa represent the petrologic type and those on the ordinate are wt% of the oxides. Data sources are in Appendix II.

YAMA et al., 1980) and the estimation for the igneous glass not to devitrify (IKEDA and TAKEDA, 1979), chondrules must have been at above 1000°C for less than several tens minutes, perhaps less than several minutes. The diffusion coefficient is relatively large at high temperatures. Consequently, if the chondrules were attached by the iron-rich rim (oxidized iron or metallic iron because troilite can not exist at above 1000°C), and if the oxidation state of the circumstance had changed, the chondrules might have become rich in FeO during a relatively short period.

#### 3. "Metamorphism" of Chondrites

#### 3.1. From the "unequilibrated" to the "equilibrated" chondrites

As mentioned above, two opposite views have been proposed on the nature of the thermal metamorphism. In general, the "unequilibrated" chondrites are characterized by the heterogeneity of the chemical composition of silicate minerals (mainly olivine and low-Ca pyroxene) and the existence of igneous glass. On the other hand, the "equilibrated" chondrites are characterized by the homogeneity of the composition of silicate minerals and the existence of sodic plagioclase (VAN SCHMUS and WOOD, 1967).

If the "equilibrated" chondrites were derived from the "unequilibrated" chondrites, the minimum time to homogenize the zoned olivine and pyroxene in the "unequilibrated" chondrites can be calculated. Recrystallization should proceed by solid diffusion, which depends on both the duration of heating and the maximum temperature. According to the Fick's law of diffusion, the diffused distance where the composition is intermediate between the core and the rim is given by the following simple equation:

$$x_{1/2} = \sqrt{Dt}$$

where x is the diffused distance, D is the diffusion coefficient, and t is the time.

Supposing the size of grain is 100  $\mu$  (average of olivine grains in ALHA77015 chondrite),  $x_{1/2}$  (distance where the composition is intermediate between the core and the rim) is 15  $\mu^*$ , and the diffusion coefficient of Mg-Fe is  $10^{-12}-10^{-14}$  cm<sup>2</sup>/s in order at 1200–900°C for Fo<sub>90-70</sub> after MISNER (1977) and TAKAHASHI (1980), then the time which is needed to homogenize zoned olivine grains is calculated as several days to several years. The diffusion coefficient of Mg-Fe in pyroxene is less than that of olivine at the same temperature, and that of Ca is much less (TAKAHASHI, 1980; MIYAMOTO and TAKEDA, 1977), so that the duration of homogenizing pyroxene grains should have been longer than the above estimation, perhaps longer by one or two orders of magnitude. In any case, the "unequilibrated" chondrites could have equilibrated within fairly short periods only by the solid diffusion.

<sup>\*</sup> This is the average of about one hundred grains which are within the porphyritic chondrules and whose zonal patterns were investigated.

# 3.2. Chemical compositions of minerals in the ordinary chondrites

The texture of the "equilibrated" chondrites varies remarkably; some of them contain chondrules with clear outlines and their matrix is fine-grained, whereas some show well recrystallized texture and the outline of chondrules are hardly defined. In spite of the variety of texture, the chemical compositions of olivine and pyroxenes are uniform and the degree of homogenization of them seems to be independent of the textural variation. Figs. 4 and 5 show the compositional variations of olivine



Fig. 4. Compositional variation of olivine in nine ordinary chondrites. One square denotes one point of measurement.

Fig. 5. Compositional variation of pyroxenes in eight ordinary chondrites.

and pyroxenes in eight ordinary chondrites. All the samples show nearly equal degree of homogenization and similar compositions. Microprobe analyses were made on the grains within the chondrules and on the grains in the matrix, and both on the core and rim of a single grain. There is no systematic compositional variation among them, and all the samples show deviations within the range of 2 to 3 moles of Fo in olivine and 1 to 4 moles of En in pyroxenes.

Those samples are considerably different in appearance; *i.e.*, Yamato-74155 contains well-defined chondrules and partly devitrified glass, whereas Yamato-74190 shows a highly recrystallized holocrystalline texture and the chondrules can not be defined\*. In spite of these textural differences, the degree of homogenization of olivine and pyroxenes is nearly the same. Consequently, olivine and pyroxenes must have been homogenized before the complete recrystallization of the chondrules.

Fig. 6 shows the compositional variation of chromite in the same samples. The



Fig. 6. Compositional variation of chromite in eight ordinary chondrites.

<sup>\*</sup> Some of the textures, such as barred olivine and radial pyroxene, are rather well preserved, even if the recrystallization of the overall texture is strong, but some, such as porphyritic and granular, are not.

compositions were calculated assuming the stoichiometry. As shown in Fig. 6, the compositional variation within a chondrite is narrow, but the variation among different chondrites is fairly large, which seems to be due to the difference of initial Cr content in the chondrites.



Fig. 7. Compositional variation of plagioclase in eight ordinary chondrites.

In contrast to olivine, pyroxenes and chromite, plagioclase shows a wide range of compositional variation. Crystallinity of plagioclase has been thought to be an indicator of the metamorphism (VAN SCHMUS and WOOD, 1967), but it does not necessarily correspond to the degree of recrystallization. In Fig. 7, four L6 chondrites show nearly the same composition of plagioclase and the range of variation is narrow. On the other hand, three H chondrites and one LL chondrite show considerably wide compositional variation of plagioclase. Though ALHA77148 belongs to H6 (MASON, 1979), the compositional variation of plagioclase is relatively large. Consequently, composition of plagioclase may not correspond to the recrystallization of chondrite.

# 3.3. "Equilibrium" temperature of the ordinary chondrites

As mentioned above, olivine, orthopyroxene, clinopyroxene and chromite in the ordinary chondrites have nearly uniform compositions, and the equilibrium relations and the equilibrium temperatures can be estimated.

Figs. 8–10 show the Fe-Mg distribution for orthopyroxene-clinopyroxene, olivine-clinopyroxene, and olivine-chromite pairs for nine chondrites. The figures indicate that the equilibrium has been attained and all the samples show nearly the same equilibrium temperature, using the geothermometers by KRETZ (1963), WOOD and BANNO (1973), WELLS (1977) and MORI (1978) for orthopyroxene-clinopyroxene pairs, by OBATA *et al.* (1974) for olivine-clinopyroxene pairs, and by EVANS and FROST (1974) and ROEDER *et al.* (1979) for olivine-chromite pairs.

The results are shown in Table 1. Pyroxenes have been equilibrated down to 900-800°C, olivine and clinopyroxene to 800-700°C and olivine and chromite to 800-





- $\Box$ : Yamato-74115,  $\triangle$ : Yamato-74155,
- ○: ALHA77148, ▲: Yamato-74007,
- ▼: Yamato-74190, ●: Yamato-74354,
- ■: Yamato-74362, +: Yamato-74646.



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Fig. 10. Relation of Fe/Fe+Mg ratios among coexisting olivine and chromite pairs. The symbols are the same as those in Fig. 8. The dotted lines represent the empirical equilibrium temperature in °C after EVANS and FROST (1975).

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		Opx-Cpx (Mg-Fe)				Ol-Cpx (Mg-Fe)	Ol-Sp (Mg-Fe)	
		Kretz	Wood and Banno	WELLS	Mori	Obata et al.	Evans and Frost	ROEDER et al.
Н	Yamato-74115	810	850	770	1110	700	730	520
	ALHA77148	930	1050	940	1000	630	750	510
	Yamato-74155	1050	1050	1020	1230	830	870	640
L	Yamato-74354	870	920	980	1000	700	750	510
	Yamato-74007	1010	900	910	990	750	880	670
	Yamato-74362	830	900	860	890	680	650	750
	Yamato-74445	1050	910	870	760	920	710	510
	Yamato-74190	870	920	880	960	650	750	540
LL	Yamato-74646	920	940	920	1040	850	810	610

Table 1. Equilibrium temperatures of ordinary chondrites.

 $700^{\circ}$ C. Diffusion of Ca in pyroxene appears to have ceased at a higher temperature than that of Mg and Fe in olivine, pyroxenes and chromite. This is obviously due to the difference in diffusion coefficient of Ca and Mg-Fe in minerals.

# 4. Metal Grains in the Ordinary Chondrites

As is well known, metals in the ordinary chondrites show a remarkable compositional zoning similar to that in iron meteorites. Analyses of the zonal pattern



Fig. 11. Frequency distribution of Ni-Fe metal grains in the ordinary chondrites. The numbers on the ordinate are those analyzed for their bulk chemical composition by the broad beam of the EPMA. The total number of a diagram represents that of metal grains in a thin section. The left side for H chondrites, and the right side for L chondrites.

reveal that the zoning was formed in situ (WOOD, 1967; NAGAHARA, 1979b).

In addition to the compositional zoning, kamacite and taenite coexist in various ways; they exist as isolated grains, contact with each other by a sharp boundary, or are associated irregularly with each other.

NAGAHARA (1979b) pointed out that the occurrence of kamacite and taenite is different between H and L chondrites: in the H chondrites, isolated kamacite and taenite are abundant and coexisting grains are less, whereas in the L chondrites, coexisting grains are abundant and isolated grains are less.

Fig. 11 shows the frequency distribution of metal grains against their wt% of Ni in five L and five H chondrites. One measurement represents the bulk composition of a single grain, where the compositional zoning, size of the grain and the coexistence of the two phases are neglected. The figure reveals that the occurrence of two phases is different between H and L chondrites. H chondrites have two distinct peaks; one is at the kamacite composition (Ni 4–7 wt%), and the other is at the taenite composition (Ni 30–40 wt%). A small peak at Ni 8–18 wt%, which is close to the average Ni content of metal in H chondrites, indicates the association of two phases in a single grain. The equilibrium temperature at which taenite of Ni 30–40 wt% and kamacite of Ni 6 wt% can coexist is about 500–400°C according to the Ni-Fe binary phase diagram (GOLDSTEIN and OGILVIE, 1965).

L chondrites have a broad peak at Ni 12–20 wt% and small peaks both at lower Ni (5–8 wt%) and at higher Ni (35–40 wt%) compositions. Metals with Ni 12–20 wt%, which is close to the average Ni content of metal in L chondrites, are mixtures of the two phases. The equilibrium temperature at which taenite of Ni 35–40 wt% and kamacite of Ni 6 wt% can coexist is about 480–450°C.

ALHA77230 exclusively shows an abnormal pattern in Fig. 11, which is rather similar to that of H chondrites. This may be due to the fact that the composition of metals is relatively rich in Fe than those in other L chondrites, or that this chondrite was cooled much slower than other L chondrites and the separation of the two phases was advanced.

The difference in the mode of occurrence of metals between H and L chondrites is considered to be due to the fact that the original composition of metals was rich in Ni in L chondrites than in H chondrites, so that kamacite began to crystallize at a higher temperature in H chondrites than in L chondrites.

Iron meteorites with Ni 8-12 wt% are coarse to medium octahedrite, which show the clear Widmastätten structure, and those with 15-20 wt% are fine octahedrite or plessite. Similarly, metals in H chondrites could tend to separate into two coarse phases, and those in L chondrites into two fine phases. Hence, if the metals in H and L chondrites cooled in the same way, the occurrence could be different.

#### 5. Discussion

# 5.1. On the nature of metamorphism

In Table 1, equilibrium temperatures for eight chondrites are listed. As the chondrites which belong to the same chemical group have a nearly constant Fe/Mg ratio, estimated equilibrium temperatures are nearly the same.

Among the H chondrites, Yamato-74155, which contains well-defined condrules and devitrified glass, shows the highest temperature. Yamato-74115, in which chondrules are considerably well defined but the degree of recrystallization is higher than the former, shows the lowest equilibrium temperature. ALHA77148 shows the intermediate degree of recrystallization and equilibrium temperature between them. The fact that the highly recrystallized chondrites show lower equilibrium temperatures and the weakly recrystallized chondrites show higher temperatures can be understood as the former had been equilibrated down to lower temperatures, while the latter to higher temperatures. This is possible if the highly recrystallized chondrites had cooled slowly and the weakly recrystallized ones rapidly.

The difference of cooling rate can be correlated to the difference of the position in the parental body from which the chondrites were derived. Chondrites which existed in a relatively inner part of the body should have cooled slowly, whereas those existed in a relatively outer part should have cooled rapidly.

If it is the case, the chondrites of higher petrologic type represent slowly cooled ones derived from the inner part of the parental body. This conclusion may support the concept of "progressive" metamorphism. However, the possibility of "autometamorphism" can not be denied, because it could also have provided the similar conditions.

In the L chondrites, no systematic difference of equilibrium temperature among the samples is found, and the deviations seem to be within the uncertainty of geothermometers. In Yamato-74354 the internal texture of chondrules is preserved and recrystallization of the matrix is weak, Yamato-74190 is intensely recrystallized, and the others show the intermediate degree of recrystallization between the former two chondrites. In spite of such a textural difference, these show nearly equal equilibrium temperature. This fact may be understood if Yamato-74354 is more recrystallized than Yamato-74115, that is, if the difference in the degree of recrystallization among those five L chondrites is smaller than that among three H chondrites.

#### 5.2. Thermal history of the chondrites

There are many conflicting models on the formation of chondrules and chondrites. It is necessary to know the relation between time(t) and temperature(T) from the beginning of the formation of chondrules to the final state of the parental body.

Here, the formation of the chondrites is thought as follows: (1) melting of preexisting materials, (2) crystallization of silicate droplets to form chondrules, (3)



Fig. 12. Equilibrium relations with decreasing temperature among the minerals. Olivine, orthopyroxene, clinopyroxene and chromite have been in equilibrium down to about 700°C as to Fe and Mg, pyroxenes as to Ca down to about 850°C. Fe-Ni metal begins to separate into two phases at about 750°C and formed the compositional zoning down to about 500°C and exsolution at about 300°C. Fe-Ni metals and troilite (FeS) have been in equilibrium down to about 300°C. Olivine and metal might have changed Fe and Ni down to 300°C, but this is inconsistent with the other elements.



Fig. 13. Schematic history of chondrites in the relation between time and temperature. Presence of core in the parental body is uncertain.

attachment of the rim and oxidation of metals with enrichement of Fe in chondrules, (4) accretion of chondrules, fragments, metals and/or troilite to form a parental body (or bodies), (5) metamorphism in the parental body to form "equilibrated" chondrites in a relatively inner part of the body and "unequilibrated" chondrites at or near the surface.

Equilibrium relations with decreasing temperature among the minerals are shown in Fig. 12 and a schematic thermal history of the "equilibrated" chondrites is shown in Fig. 13.

The mechanisms of melting of the pre-existing materials and crystallization of melt droplets are not discussed in this paper. They are discussed by IKEDA and TAKEDA (1979) and TSUCHIYAMA *et al.* (1980). The cooling rate of chondrules must have been 50-120 deg/min and they were kept above  $1000^{\circ}$ C for less than several minutes, when the enrichment of Fe occurred near the surface of some chondrules.

It is questionable whether the accretion took place immediately after the formation of the chondrules or far after. This problem relates to the mechanism of formation of the chondrules. If the chondrules were formed by impact of the bodies (KIEFFER, 1975; MARTIN and MILLS, 1976; KERRIDGE and KIEFFER, 1977), the dispersed chondrules might have accreted immediately, but if they were formed by another mechanism such as discharge or direct condensation from the nebula, they did not necessarily accrete immediately.

The accreted chondrules, fragments, fine materials, metals and troilite formed chondrites. The energy of the accretion might have caused at least sintering; that is, the loose materials became tight chondrites. Furthermore, the "auto-metamorphism" might have occurred. As discussed above, it is not well understood how the recrystallization occurred. It might be caused by reheating due to the decay of the radioisotopes or the accretion energy.

During the reheating, chondrites should have been heated at least to the "equilibrium" temperature, about 900–1000°C, and the heterogeneous minerals (olivine and pyroxenes) could have been become homogeneous in a relatively short period. Chondrites which existed in a relatively inner part of the body could preserve equilibrium relations among minerals down to about 800°C and the matrix was well recrystallized, while those which existed in the outer part were cooled relatively rapidly, resulting in the high apparent equilibrium temperature and weak recrystallization.

Down to about 700°C, olivine and pyroxenes could preserve the equilibrium relations, and at or below 700°C, Ni-Fe metal began to separate into two phases. Down to 500–400°C, the separation continued and the compositional zoning was formed at lower temperatures. With further decrease of temperatures down to 300°C, the exsolution of kamacite in the taenite grains occurred, which was the lowest temperature recorded in the chondrites.

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## APPENDIX

#### I. Brief petrography of the samples

(1) Yamato-74007 (L6)

Mildly recrystallized texture. Outline of the chondrules is obscure and their internal texture is barely preserved. Fine grained plagioclase is present.

(2) Yamato-74445 (L6)

Mildly to severely recrystallized texture. Outline and internal texture of the chondrules are obscure. Plagioclase is barely observed.

(3) Yamato-74155 (H4)

Weakly recrystallized texture. Outline and internal texture of the chondrules are well preserved. Polysynthetic twinned pyroxene and low-Ca pyroxene are present.

(4) ALHA77148 (L6)

Texture of chondrules is well preserved. Their outline is obscure. Matrix is weakly recrystallized. Plagioclase is present. Olivine and pyroxenes have uniform compositions.

(5) ALHA77015 (L3)

It consists of closely packed chondrules with a minor amount of the fine-grained matrix. Olivine and pyroxene show wide compositional variation.

(6) ALHA77033 (LL3)

It consists of closely packed chondrules and fragments of them with a small amount of the fine-grained matrix. Olivine and pyroxene show compositional variations.

# II. List of chondrites compiled in Fig. 3

See "Cambridge Chondrite Compendium" (MOTYLEWSKI, 1977) for each data source and classification.

- H 3 Bremervöide, Clovis, Sharps, Tieschity
- H 4 Ankober, Beaver Creak, Guarena, Kaba, Kiffa, Pulsora, Schenectady, Weston
- H 5 Allegan, Andura, Castallia, Collescipoli, Cronstad, Dwaleni, Forest City, Geidam, Gnadenfrei, Hessle, Knyahinya, Salt Lake City, Sitathali, Stalldalen
- H 6 Benld, Butsura, Cape Girardeau, Cobija, Djati-Pengilion, Estacado, Kernouve, Kimble County, Mount Browne, Oakley, Plantersville
- L 3 Bishunpur, Bovedy, Carraweena, Hallingerberg, Ioka, Krymka, Mabwe Khoywa, Manych, Mezö-Madaras
- L 4 Albareto, Atarra, Barratta, Bjurböle, Cynthiana, Goodland, McKinney, Rupota, Saratov, Tennasilm
- L 5 Chandakapur, Crumbin, Hendersonville, Homestead, Malakal, Mirzapur, Mooresfort, Shelburne, Wethensfield, Witterkranz
- L 6 Baroti, Chantonnay, Coon Butte, Cranganore, Forksville, Girgenti, Grossliebenthal, Harleton, Karth, Kuttippuram, Launton, Lesves, Lissa, Lundsgard, Mauerkichen, Modoc, Orvinio, Perpeti, Rakovka, Rich Mountain, Segowlie, St. Michel, Sultanpur, Tathlith, Tourinnes-la-Grosse, Varpaisjarvi, Zemaitkiemis
- LL 3 Chainpur, Ngawi, Parnalee, Semarkona
- LL 4 Hamlet, Soko-Banja
- LL 5 Cherokee Springs, Olivenza
- LL 6 Appley Bridge, Douan Mghila, Ensisheim, Jelica, Manbhoom, Mangwendi, Näs, Ottawa, St. Séverin, Vavilovka