

Minerals in Volcanic Ash 2: Non-magmatic Minerals

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Abstract

Non-magmatic minerals are abundant in volcanic ash, especially in volcanic ash of phreatic and phreatomagmatic eruptions. Accidentally incorporated minerals from country rocks (rock into which magma or water intrude) are common in all types of eruptions. Any kind of rock-forming minerals in the crust and mantle may be incorporated into volcanic ash by mechanical stripping from conduits or magma chambers. Volcanic ash often contains hydrothermal minerals, such as smectite, kaolinite, cristobalite and gypsum, that are formed by rock-fluid or magma-fluid interactions. Hydrothermal minerals are common in the volcanic ash of phreatomagmatic and phreatic eruptions. Some kinds of minerals in volcanic ash must have precipitated from volcanic fluids that involve hydrothermal water in subvolcanic aquifers, crater lakes, fumarolic gases, and volcanic gases. Sulfate, halide, and native sulfur may be the major minerals precipitating from volcanic fluid, but they have not been well documented.

Volcanic ash commonly contains hydrothermal clay minerals which are similar to soil clay formed by weathering of magmatic fragments. Sulfide and halide minerals in volcanic ash have an influence on human environments such as through soil properties and climate.

Key words: hydrothermal minerals, leachate, magma-water interaction, non-magmatic minerals, rock-water interaction, xenocrysts

1. Introduction

Volcanic ash contains many kinds of non-magmatic minerals. We use here the term “magmatic minerals” for the minerals crystallized from magma, or for common rock-forming minerals in igneous rocks, and “non-magmatic minerals” for all the other minerals in volcanic ash. So far little attention has been given to non-magmatic minerals in volcanic ash because volcanologists have mainly focused on magmatic minerals. Studying magmatic minerals seems essential to understanding volcanic activities since their features sensitively reflect physical and chemical conditions of magmas. On the other hand, non-magmatic minerals have been considered of less importance because they are only accidental or secondarily-produced materials.

In other research fields on volcanoes aside from mineralogy and petrology, external conditions outside the magma have been long studied. Interactions

among volcanic gases, hydrothermal water, country rock, and magma have been studied by researchers of volcanic gas, water, geothermics, and economic geology (e.g., Giggenbach, 1997). During the last two decades, some geologists have discussed magma-water interactions that cause explosive eruptions (Wholetz and Heiken, 1992; Wholetz, 2002, 1986; Zimanowski *et al.*, 1997; White, 1996; Barberi, 1985; etc.). Non-magmatic minerals are produced through those interactions, and incorporated into volcanic ash of explosive eruptions. Those minerals appear to be clues to understanding the interactions between magmas and the external circumstances (Heiken and Wohletz, 1992). Even those non-magmatic minerals derived from country rocks (rock into which magma or water intrude) without any interaction give us valuable information such as depths of explosions. In this report, we will describe the types and origins of non-magmatic minerals in volcanic ash from the viewpoint of interaction among

magma, water, gas, and country rocks.

In spite of little attention to non-magmatic minerals, they are of great importance to environmental science. Non-magmatic minerals contain some elements, such as sulfur or halogens, that are essential for the environment. Such minerals are more water soluble than magmatic minerals, hence, their components easily enter environmental waters. There are also clay minerals, such as kaolinite, which influence soil characteristics. Thus the influence of non-magmatic minerals on the environment should be studied in greater depth.

2. Non-magmatic-mineral-producing Eruptions

Most volcanic eruptions discharge some amount of non-magmatic minerals. The abundance of non-magmatic minerals in volcanic ash correlates to eruption styles in terms of magmatic, phreatomagmatic, and phreatic eruptions. Volcanic ash of magmatic eruptions is composed of abundant magmatic materials (volcanic glass and magmatic minerals) and a small amount non-magmatic materials (non-magmatic minerals and rock fragments), whereas that of phreatic eruptions consists only of non-magmatic materials. Constituents of phreatomagmatic eruption products show a wide range of ratios between magmatic and non-magmatic materials.

Explosive magmatic eruptions are caused by degassing and bubbling under the condition of volatile-saturation by decompression. Since explosive fragmentation occurs within the magma itself and since the explosivity is lower than for other types of eruptions, non-magmatic materials from external circumstances are not necessarily contained in volcanic ash. Nevertheless, non-magmatic minerals are often contained in magmatic volcanic ash such as "xenocrysts," oxidized iron-oxide minerals, sulfates, and sulfides. "Xenocryst" is the term used for accidentally incorporated minerals from country rock into magma. Xenocrysts are derived from various depths from the mantle to the surface. Reddish-coloured scoria contains hematite and/or goethite that could be regarded as non-magmatic minerals. Those minerals are secondarily produced by oxidation or hydration during eruptions. Sulfate and sulfide minerals are not usually regarded as magmatic minerals, but both minerals were contained in the volcanic ash from eruptions of Mt. Pinatubo in 1991 (Hattori, 1997) and El Chihcon in 1982 (Luhr *et al.*, 1984). Since the origin of sulfate and sulfide minerals in magma is arguable, we will not discriminate here between magmatic and non-magmatic minerals.

Phreatic eruptions, or steam explosions, are caused by the abrupt expansion of steam derived from a sub-volcanic aquifer. Sudden steam expansion is caused

by decompression and/or heating of hydrothermal fluid. Since magma is not discharged by the eruptions, the volcanic ash consists only of non-magmatic materials. Non-magmatic materials in phreatic volcanic ash are derived mainly from subsurface country rocks. Explosions caused by sudden steam expansion result in fragmentation of country rocks with high explosivity. Volcanic ash from phreatic eruptions also contains considerable amounts of minerals produced by hydrothermal alteration. Before a phreatic eruption, country rocks under the crater may have reacted with the hydrothermal fluid that is possibly essential for steam expansion during the phreatic eruption. Some minerals may precipitate directly from hydrothermal water. The mineral properties reflect hydrothermal water chemistry, temperature, and pressure, hence, properties of the fluid may be estimated from the mineralogy of the volcanic ash.

Phreatomagmatic eruptions are caused by magma-water interaction such as magma intrusion into subvolcanic aquifers or crater lakes. The explosive mechanism of magma-water interaction, e.g., the MFCI mechanism (Zimanowski, 1997), has been a subject of controversy over the last few years. High explosivity and dynamics of magma-water interaction are of great interest to modern volcanology. Volcanic ash from the eruptions shows a wide range of non-magmatic vs. magmatic mineral ratios. If steam and magma are the only materials discharged by a phreatomagmatic eruption, the volcanic ash consists only of magmatic materials (glass and magmatic minerals). However, volcanic ash of phreatomagmatic eruptions usually contains considerable amounts of non-magmatic materials. Some phreatomagmatic eruptions discharge only a trace of magmatic minerals. For example, volcanic ash from the phreatomagmatic eruption at Mt. Usu on 31 March 2000 contains trace amounts of magmatic minerals and glass shards. Some of the non-magmatic minerals in phreatomagmatic volcanic ash originate from water-magma interaction, in addition to country rock and hydrothermal water.

3. Minerals in Volcanic Ash

3.1 Xenocrysts in magmatic and phreatomagmatic volcanic ash

Xenocrysts occur both as free crystals and in magmatic fragments (glass shards, pumice, and scoria) in magmatic- and phreatomagmatic volcanic ash. Although any mineral may be incorporated into magma, minerals recognized as xenocryst are limited to only a few tens of mineral species, since most solid parts of the Earth are composed only of small numbers of minerals called "rock-forming minerals." Quartz, feldspars, olivine, pyroxenes, amphiboles, and micas commonly occur as xenocrysts, but these minerals are often indistinguishable from phenocrysts because they also commonly occur as phenocrysts. These xeno-

crysts often exhibit peculiar textures (kink band, corroded form, reaction rim, breakdown aggregations) that help to discriminate them from phenocrysts. Phenocryst mineral assemblages can be roughly estimated from magma compositions that are provided by chemical analysis of glass shards, pumice or scoria. Therefore, we can recognize xenocrysts if some minerals in the magmatic fragments are unlikely to be in equilibrium with the magma.

Quartz often occurs as xenocrysts in magmatic fragments in which quartz is not equilibrated, such as basalt, andesite, and alkaline rock fragments. Quartz xenocrysts exhibit corroded forms caused by resorption in magma, or reaction rims caused by mineral crystallization after quartz-magma reactions (Sato, 1975). Plagioclase xenocrysts also exhibit resorption textures that are called spongy texture, sieved texture, or dusty zones. Resorption textures of these minerals are caused not only by incorporation from country rocks but also by mixing of two or more magmas. Careful petrological observation is needed to distinguish xenocrysts from mixing disequilibrium. Hornblende and biotite also occur as xenocrysts. When hornblende or biotite is brought to the surface by hot magma, the mineral changes into an aggregation of other minerals (pyroxene, feldspar, iron oxide), which is called "breakdown." Since breakdown texture is also formed by magma mixing, careful observation is needed to discriminate xenocrysts from magma mixing.

Xenocrysts of olivine and pyroxenes are often interpreted as having a mantle-peridotite origin. Those minerals are the major constituents of peridotite in the upper mantle where basaltic magmas are generated. Peridotite fragments, called "mantle xenolith," sometimes occur in volcanic ash, e.g., the Ichinomegata volcano, Oga Peninsula, NE Japan. Minerals disaggregated from mantle xenoliths are xenocrysts, but they are sometimes confused with phenocrysts if there are no signs of xenocrysts such as a kink band that is a sharp bending plane in a crystal. Xenocrysts from crustal rocks show a wider variety than those from mantle rocks, since crustal rocks comprise larger numbers of rock-forming mineral species than mantle rocks, which consist of only a few kinds of minerals. Major crustal minerals are quartz, feldspars, pyroxenes, olivine, hornblendes, micas, garnet, aluminosilicates, carbonates, and Fe-Ti oxide. The mineralogy of xenocrysts from crustal rocks sometimes becomes a good tool for estimating crustal features (geothermal gradient, composition, etc.). For example, Hacker *et al.* (2000) estimated the geothermal gradient and development history of the Tibetan Plateau from the mineralogy of xenocrysts. Xenocrysts also give some clues for evaluating thermal interaction between magma and the crust. Barberi and Leoni (1980) discussed thermal interaction between a shallow magma chamber and crustal carbonate rocks based on the mineralogy of carbonate

xenocrysts in the volcanic ash from Vesuvius.

3.2 Hydrothermal-alteration-related minerals in phreatic and phreatomagmatic volcanic ash

Volcanic ash of phreatic and phreatomagmatic eruptions commonly contains considerable amounts of minerals produced by hydrothermal alteration prior to the eruptions. Hydrothermal minerals, such as smectite, kaolinite and cristobalite, commonly occur in these volcanic ashes. Occurrence of the the same kinds of minerals have been well documented in active geothermal fields (Hayashi, 1973; Browne, 1978; Henley and Ellis, 1983) and around hydrothermal ore deposits (e.g., Reed, 1997; Izawa, 1991), whereas they have been less described for volcanic ash (Ossaka and Ozawa, 1966; Kudo *et al.*, 2000). Since all these geological environments relate to volcanic-hydrothermal systems, occurrence of these minerals must be similar in each of these environments. According to studies on geothermal fields and hydrothermal ore deposits, hydrothermal mineral assemblages strongly depend on both the acidity and temperature of the hydrothermal water. Therefore, hydrothermal mineral assemblages in volcanic ash must reflect physical and chemical properties of the hydrothermal water. Some workers previously presented schemes of spatial chemical variation of hydrothermal water beneath volcanic edifices (e.g., Browne, 1978; Hayashi, 1978; Henly and Ellis, 1983). Compared with the schemes, hydrothermal minerals in volcanic ash will appear in their original position in the volcanic edifice and have their original properties as well. Descriptions of hydrothermal minerals in volcanic ash have been limited, but we list some description by recent researchers in Table 1. We will report an example of hydrothermal minerals from the volcanic ash of the 1997 eruptions of the Akitayakeyama volcano. The volcano erupted twice that year; first on 11 May at the foot of the stratocone and second on 16 August at the summit crater. Mineral assemblages of the volcanic ash differ between the two eruptions, as shown in Table 2. Compared with the mineralogical works in geothermal fields (Browne, 1978; Hayashi, 1973; Henley and Ellis, 1983), hydrothermal minerals in the volcanic ash of 11 May appear to be equilibrated with hydrothermal water of low temperature (<100°C) and their acidity is neutral to weakly alkaline. On the other hand, those of 16 August were equilibrated with high-temperature and acidic hydrothermal water. Detailed observation of the minerals in these volcanic ashes from Akitayakeyama gives us a concrete picture of subvolcanic hydrothermal systems. Itoh *et al.* (1997) estimated the source depth of the phreatic eruption on 11 May based on a comparison between the volcanic ash and drill holes. The presence of heulandite (zeolite) in the volcanic ash indicates alkaline hydrothermal water at a shallow depth, which is

consistent with the scheme of subvolcanic water chemistry that involves alkaline hydrothermal water

shards. Palagonite is not a true mineral name but a mixture of fine glass, smectite, iron oxide, and other

Table 1 Hydrothermal minerals in phreatic eruption deposits.

Volcanoes	Ages	Minerals	References
Jogokunuma-Hakkoda	500-300y .B.P.	crystalobalite, tridymite, quartz, smectite, kaolinite	Kudo <i>et al.</i> (2000)
Iwate	500-100y .B.P.	kaolinite, cristobalite, alunite, montmorillonite, pyrophyllite, quartz	Itoh (1999)
Bandai	1888	kaolinite, illite/smectite mixed layer	Yamamoto <i>et al.</i> (1999)
Yakedake	1962	montmorillonite, mica, quartz, cristobalite	Ossaka and Ozawa (1966)
Kuju	1995	cristobalite, quartz, alunite, pyrite, gypsum, kaolinite, anatase, native sulfur	Taguchi <i>et al.</i> (1996)
Hokkaido-komagatake	1996	montmorillonite	Ui <i>et al.</i> (1996)
Akita-Yakeyama	1997	See Table 2	Itoh <i>et al.</i> (1997)
Usu	2000	quartz, smectite, chlorite, calcite, pyrite, zeolite	Website of Geological Survey of Hokkaido

Table 2 Hydrothermal mineral assemblages in volcanic ash of phreatic eruptions of the Akita-Yakeyama volcano in 1997.

Date	11 May 1997	16 August 1997
location	northeastern flank	summit crater
minerals	montmorillonite, kaolinite, tridymite, quartz, pyrite, heulandite, Yugawalite (Itoh, <i>et al.</i> 1997)	montmorillonite, pyrophyllite, kaolinite, gypsum, cristobalite, quartz, alunite, pyrite, anatase, diaspore, andalusite
estimated depth	< 100 m (Itoh <i>et al.</i> , 1997)	0 - 1000 m
estimated temperature	<100°C	< 330°C
hydrothermal water acidity	acid – weak alkaline	intense acid - weak acid

near the foot of the volcano (Henley and Ellis, 1983). On the other hand, minerals in volcanic ash from the summit crater indicate the core of the volcanic edifice as the source region. Quartz aggregation is quite abundant in the ash, which means there is a “silica baggy” (Giggenbach, 1997) that is formed by leaching of most elements by acidic water at the core of the volcanic edifice. The presence of sulfides (pyrite) and sulfates (gypsum and alunite) indicates a high-sulfidation environment. Paragenesis of pyrophyllite + quartz and andalusite + quartz in altered rock fragments indicates hydrothermal conditions of high temperature (230-330°C) and high acidity. Since water cannot exist in liquid form under such high temperatures or at low pressure conditions, high pressure conditions at a depth of hundreds of metres would be needed to crystallize the minerals.

3.3 Magma/water-interaction-related minerals in phreatomagmatic volcanic ash

It is well known that direct contact between magma and water induces explosions, and it is also known that some minerals are produced by the reaction between magma and water at the time of contact. “Palagonite” is the term used for the material that is considered to be produced by direct interaction between magma and water. Palagonite usually forms on the surface of lava, pumice, scoria, and glass

minerals (Heiken and Wohletz, 1992).

Hematite (Fe₂O₃) and goethite (FeOOH) are not common magmatic minerals but are produced in magmatic fragments by oxidation and/or hydration through water-magma or air-magma interactions. Reddish-coloured magmatic fragments (scoria, pumice, glass shards) contain those iron-oxide minerals that are formed by oxidation or hydration of iron in volcanic glass or magnetite.

3.4 Other non-silicate minerals in magma

Except for a couple of Fe-Ti oxide minerals (magnetite and ilmenite), zircon, and apatite, non-silicate minerals in magmatic fragments have not been well described regardless of whether they are magmatic or non-magmatic. Some kinds of oxide, sulfide, and sulfate minerals are considered to be key minerals in understanding the behavior of magma. We will discuss here those non-silicate minerals even though some of them might be magmatic minerals.

Spinel and chromite in volcanic ash might be magmatic minerals, in particular, in basaltic magmas. But they also might be xenocrysts especially from mantle peridotite, the same as pyroxene and olivine xenocrysts. As stated above, hematite and goethite are produced by oxidation and/or hydration of iron in magma. Sulfide and sulfate minerals, such as isocubanite or gypsum, in volcanic ash have been focus-

ed on during the last decade, since those minerals in volcanic ash from active volcanoes are considered to give an understanding of the behavior of volcanic gas and magma (Hattori, 1997). The presence of anhydrite (CaSO_4) in the volcanic ash of the eruption at Mt. Pinatubo in 1991 is vital for considering the inconsistency between excess sulfur gas emission and the sulfur budget in the magma. Similar excess sulfur emission was observed at the eruption of El Chihcon in 1982 (Luhr *et al.*, 1984). Those examples are not exceptional with regard to the presence of sulfide and sulfate minerals, even though those minerals have not been well described in magmatic fragments or volcanic rocks. Since economic geologists have recognized sulfur-bearing minerals in magmatic ore deposits, (Naldrett, 1989), those minerals seem to be common magmatic fragments, but they have been ignored by petrologists.

4. Origin of Non-magmatic Minerals

Mechanical incorporation of country rocks

Since magmas rise through the crust to the surface, any minerals along the path may be accidentally incorporated into volcanic ejecta. Minerals are stripped from the wall of the conduit by the ascending magma. Minerals incorporated by this process occur as both free crystals and xenocrysts in magmatic fragments. Non-magmatic eruptions, or phreatic eruptions, also discharge country rock fragments that come from the shallow crust. In this case, the minerals are stripped from country rocks by ascending steam or water.

Non-magmatic minerals in volcanic ash of phreatomagmatic eruptions are brought up by steam, water, and magma. Because of the wide variety of rock-forming minerals in crustal rocks, many kinds of rock-forming minerals can be incorporated by this mechanical process. Since many magmas pass through the mantle, minerals from mantle peridotite,

accidentally included in volcanic ash.

Alteration of country rock and magmas

Hydrothermal water commonly interacts with country rocks under volcanic edifices, and an area of hydrothermal alteration develops around the volcanic edifices. The interaction causes mineralization of hydrothermal minerals, then the country rocks change into altered rocks. Hydrothermal water beneath a volcano has a peculiar character because of heating by magma and injection of volcanic gas into the water, hence, species in the altered rocks around the volcano are unique. Most hydrothermal minerals in volcanic ash are derived from altered rocks that formed prior to the eruption. The process of incorporation of the minerals into volcanic ash is the same as that of country rock fragments, but the minerals are different from common rock-forming minerals in crustal and mantle rocks (Table 3).

Direct interaction between water and magma also produces such hydrothermal minerals. This mineral interaction only lasts for a short period during the eruption. Even during such a short time this interaction induces mineralization of hydrothermal minerals, but hydrothermal alteration does not penetrate all of the magma. The interaction often results in thin crusts of hydrothermal minerals (palagonite) surrounding magmatic fragments. If the instantaneous interaction results in only oxidation or hydration of the iron in the magma, the magmatic fragments change their color into red by formation of hematite or goethite.

Precipitation from water and gas

In volcanic environments, many kinds of minerals continuously precipitate from volcanic fluid (hydrothermal water in subvolcanic aquifers, crater lake water, volcanic gas, and fumarolic gas). When a volcano erupts explosively, the volcanic plume might mechanically carry those minerals that have precipi-

Table 3 Potent non-magmatic minerals in volcanic ash and their origins.

origins of minerals	potent minerals	incorporation mechanisms
crustal and mantle rocks	plagioclase, quartz, alkali-feldspar, mica (biotite, phlogopite, etc.), amphiboles, pyroxenes, olivine, garnet, Fe-Ti oxide, spinel, calcite, dolomite, etc.	mechanical stripping from country rocks by ascending magma, steam, or water
related to external water (hydrothermal alteration, water-magma reactions, precipitation from volcanic fluids)	quartz, cristobalite, tridymite, smectite (montmorillonite, saponite, etc.), sericite, illite, mixed layer, kaolinite, zeolite (mordenite, heulandite, etc.), alunite, gypsum, anhydrite, pyrophyllite, chlorite, albite, aduralia, pyrite, anatase, diaspore, native sulfur, etc.	<ul style="list-style-type: none"> • mechanical stripping from hydrothermally altered country rocks by ascending magma, steam, or water • water-magma interactions • mechanical incorporation from aquifers or surface water into plume • direct precipitation from volcanic fluids

such as olivine, pyroxenes and spinel, are also

tated prior to the eruption. Species of minerals that

precipitate from volcanic fluids are similar to those in hydrothermally altered rocks, but direct precipitation from fluid results in a wider variety of mineral species. Rare minerals are often found especially around fumaroles, such as orpiment (As_2O_3). Major minerals precipitated from volcanic fluids include quartz, cristobalite, gypsum, alunite, native sulfur, and iron oxide. These minerals must commonly occur in volcanic ash, but so far they have not been well described. Native sulfur was reported from the volcanic ash of the 1995 eruption of the Ruapehu volcano (Christenson, 2000).

Some minerals might precipitate from volcanic fluids even at the time of the eruption. During an eruption, physical conditions of volcanic fluids change dramatically, involving phase transition of fluids, decompression, and rapid cooling. Since the period of sustainment in the volcanic column is short, mineral precipitation in the volcanic plume requires a rapid crystallization rate, which is feasible for some kinds of sulfate and halide (chloride and fluoride) minerals but is unlikely for silicate minerals. If the minerals precipitate in the volcanic plume, they must account for a certain part of the volcanic ash. However, they are hardly recognized as constituents of volcanic ash, because they occur as very fine grains and because such minerals are so water soluble that rainfall can flush them away. In fact, there is no description of certain minerals that are precipitated from volcanic fluids during an eruption. That is why we did not describe such minerals in the above sections. Nevertheless, such minerals evidently precipitate from volcanic fluids in the volcanic plume. During the last two decades, the volcanological significance of "leachate," or soluble components on the surface of volcanic ash particles, has been emphasized by researchers on volcanic fluids (e.g., Casadevall, *et al.*, 1983). They consider that when volcanic ash is suspended in a plume, a certain portion of the gas phase condenses onto the surface of ash particles. They found that the condensates are soluble sulfates, chlorides, and fluorides. If the volcanic gas phase is condensed in the free space in the plume, the condensate becomes an "aerosol" and is suspended in the air for long periods.

5. Environmental Significance

No one has been able to evaluate the effects of non-magmatic minerals in volcanic ash on the human environment since insufficient descriptions of the minerals are available. We will mention here two possible effects on human environments based on the descriptions in above sections. One is effects on soil and the other is that on climate.

Properties of soils are dominated by clay minerals that are thought to be formed by weathering of magmatic fragments after deposition. Clay minerals formed by weathering of magmatic fragments include

smectite, kaolinite, mica, etc. However, as stated above, volcanic ash commonly contains those clay minerals that are formed by hydrothermal activities around volcanic edifices. Pedological effects of the primarily contained clay minerals in volcanic ash should be evaluated for distribution, properties, and the genesis of soils. Sulfate and halide minerals in volcanic ash are of great significance with regard to properties of soils. Even small amounts of soluble minerals govern chemical properties of soils since they bring some essential anions such as SO_4^{2-} , Cl^- , F^- . Penetration of these anions into fields sometimes causes disastrous effects. Fluoride-rich volcanic ash from the Hekla volcano in 1970 caused 7,000 sheep to die from feeding on polluted grass.

Volcanic ash is one source of aerosols that dominate the global climate. Magmatic and non-magmatic rock fragments may become aerosols if they are fine-grained enough. Condensate from volcanic fluids will be more significant for aerosol formation. Condensation often occurs at high levels in the volcanic plume, sometimes even in the stratosphere. Grains of the condensate are usually small enough to be suspended in the air for years and spread around the world, whereas most magmatic and rock fragments fall onto the ground around the volcano. Aerosol from the volcanic fluid discharged by the eruption effects climate cooling. This cooling can negatively affect agriculture. Furthermore, soluble components in the aerosol can increase rainfall acidity.

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