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MORPHOLOGICAL AND CHEMICAL CHARACTERIZATION OF NEUTRALLY BUOYANT PLUME-DERIVED PARTICLES AT THE EASTERN MANUS BASIN HYDROTHERMAL FIELD, PAPUA NEW GUINEA

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Abstract

Neutrally buoyant particulate matter emitted from hydrothermal plumes in the eastern Manus Basin hydrothermal field, Papua New Guinea, was detected and sampled by CTD-transmissometer surveys during marine expeditions in 1991 and 1993. Characterization of suspended particulate matter has helped to prove that the main sources of venting are along a prominent 20-km-long NE–SW-striking felsic volcanic ridge. Elemental composition and particle morphology were determined by scanning electron microscopy and X-ray energy-dispersion spectroscopy. The size distribution and relative abundance of particles were determined by image analysis on selected samples. By looking at the composition of particulate matter, we can infer the origin, maturity and proximity of the source that is producing the particles. Seven types of particles have been morphologically and chemically characterized: i) filaments of probable biogenic origin of a Mn–Fe–Si phase containing traces of Cu and Zn, ii) orb-like particles of an Fe–Mn–Si phase containing minor Mg, Al, P, K and Ca, iii) barite, iv) anhydrite, v) colloidal-sized amorphous iron oxide, vi) an unidentified Si–Fe-rich phase, and vii) widespread biogenic detritus (shrimps, diatoms, silicoflagellates, copepods and unidentified gelatinous organic matter). Most of the particles are <2 μm in size and range from <2 μm for amorphous iron oxide to 100 μm for anhydrite and the Si–Fe-rich phase. Studies on suspended particulate matter constitute an effective way to recognize hydrothermal plumes.

Keywords: hydrothermal particles, suspension, marine particles, characterization, hydrothermal plumes, Manus Basin, Papua New Guinea.

Sommaire

Nous documentons la présence de particules en suspension émanant de panaches hydrothermaux dans le champ d'activité hydrothermale de l'est du bassin de Manus, en Papouasie Nouvelle Guinée; nous avons échantillonné ce matériau lors de relevés par transmissométrie CTD au cours d'expéditions marines en 1991 et 1993. Suite à cette caractérisation des particules, nous préconisons, comme sources principales des évents, une crête orientée NE–SO longue de 20 km et faite de roches volcaniques felsiques. La composition et la morphologie des particules ont été établies par microscopie électronique à balayage et par analyse en dispersion d'énergie. La dimension et la concentration des particules ont été établies par analyse d'images de certains échantillons. La composition des particules mène à l'origine, la maturité et la proximité des sources responsables. Nous décrivons la morphologie et la composition de sept sortes de particules: i) filaments d'origine biogénique probable contenant une phase à Mn–Fe–Si et des traces de Cu et de Zn, ii) particules orbiculaires d'une phase à Fe–Mn–Si contenant un peu de Mg, Al, P, K et Ca, iii) barite, iv) anhydrite, v) oxyde de fer amorphe de dimension colloïdale, vi) une phase riche en Si et Fe non identifiée, et vii) détritus biogénique répandu (crevettes, diatomes, silicoflagellés, copepodes et matériau gélatineux non identifié). Dans la plupart des cas, les particules ne dépassent pas 2 μm et vont de <2 μm (oxyde de fer amorphe) à 100 μm (anhydrite et phase riche en Si et Fe). On démontre l'efficacité d'une étude de telles particules pour reconnaître l'existence de panaches hydrothermaux.

(Traduit par la Rédaction)

Mots-clés: particules hydrothermales, particules marines, suspension, caractérisation, panaches hydrothermaux, bassin de Manus, Papouasie Nouvelle Guinée.

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Introduction

The interaction of high-temperature fluids and ambient seawater at spreading centers along ocean ridges and back-arc basins produces fine-grained precipitates of metal-rich sulfides, sulfates and Fe and Mn oxyhydroxides (Haymond & Kastner 1981, Leinin 1985, Feely *et al.* 1987). The physical and chemical processes that govern the off-axis dispersion and composition of hydrothermal precipitates are not well understood. However, neutrally buoyant plumes are considered to be an important mechanism of dispersal for some hydrothermal species into the ocean and pelagic sediments (Craig & Lupton 1981, Edmond *et al.* 1982, Lupton *et al.* 1985, Baker & Massoth 1987, Mottl & McConachy 1990). Studies of the settling flux of hydrothermal-plume-derived particles have shown that 90% of the material ejected is transported farther than two kilometers from its source (Dymond & Roth 1988). Experimental studies of the particle fallout have shown that the finest material, which consists of 90% of the total emission, does not settle out during the first stage of the development of the plume. Rather, it is carried with the rising plume and dispersed (Foster 1982).

Particle-laden plumes undergo further alteration, such as mineralization processes (Mottl & McConachy 1990), scavenging (Cowen *et al.* 1986, 1990, Weiss 1977) and dissolution (Feely *et al.* 1987) as they spread out laterally. Mineral and chemical characterization of marine particles has been extensive for a variety of hydrothermal sites at mid-ocean ridges and sedimented basins (Baker *et al.* 1985, Feely *et al.* 1987, Trocine & Trefry 1988, Walker & Baker 1988, Mottl & McConachy 1990, Kadko *et al.* 1990, Feely *et al.* 1990, 1992, 1994a, b). The analysis has extended to particles of very diverse nature (Roth & Dymond 1989, Cowen 1991, Mottl 1991).

The newly precipitated hydrothermal material above the vent source contains particles of fresh minerals that are typically of large size before they are transported far afield. Characterization of this hydrothermal material has been done mainly on particles in the rising plume (Mottl & McConachy 1990, McConachy 1988, Feely *et al.* 1990). Detailed characterization of hydrothermal material in neutrally buoyant plumes, the subject of this study, has only rarely been attempted (Walker & Baker 1988).

Background Information

The eastern Manus Basin was surveyed in 1991 and 1993, and active hydrothermal venting was detected over the informally named Pual Ridge (Binns & Scott 1993). The PACMANUS I and II expeditions detected, by means of *in situ* measurement of physical properties (transmissivity, temperature, salinity), indications of hydrothermal activity in the water column, and sampled the suspended particulate material within neutrally buoyant plumes. Such plumes were typically traced as far as 20 km from the source. Earlier descriptions of hydrothermal particles from active sources in the eastern Manus Basin indicated their presence in neutrally buoyant plumes (Binns & Scott 1993, Ortega-Osorio & Scott 1995) in which particles reach a height of neutral buoyancy between 150 and 250 m above the sea floor before they spread out horizontally.

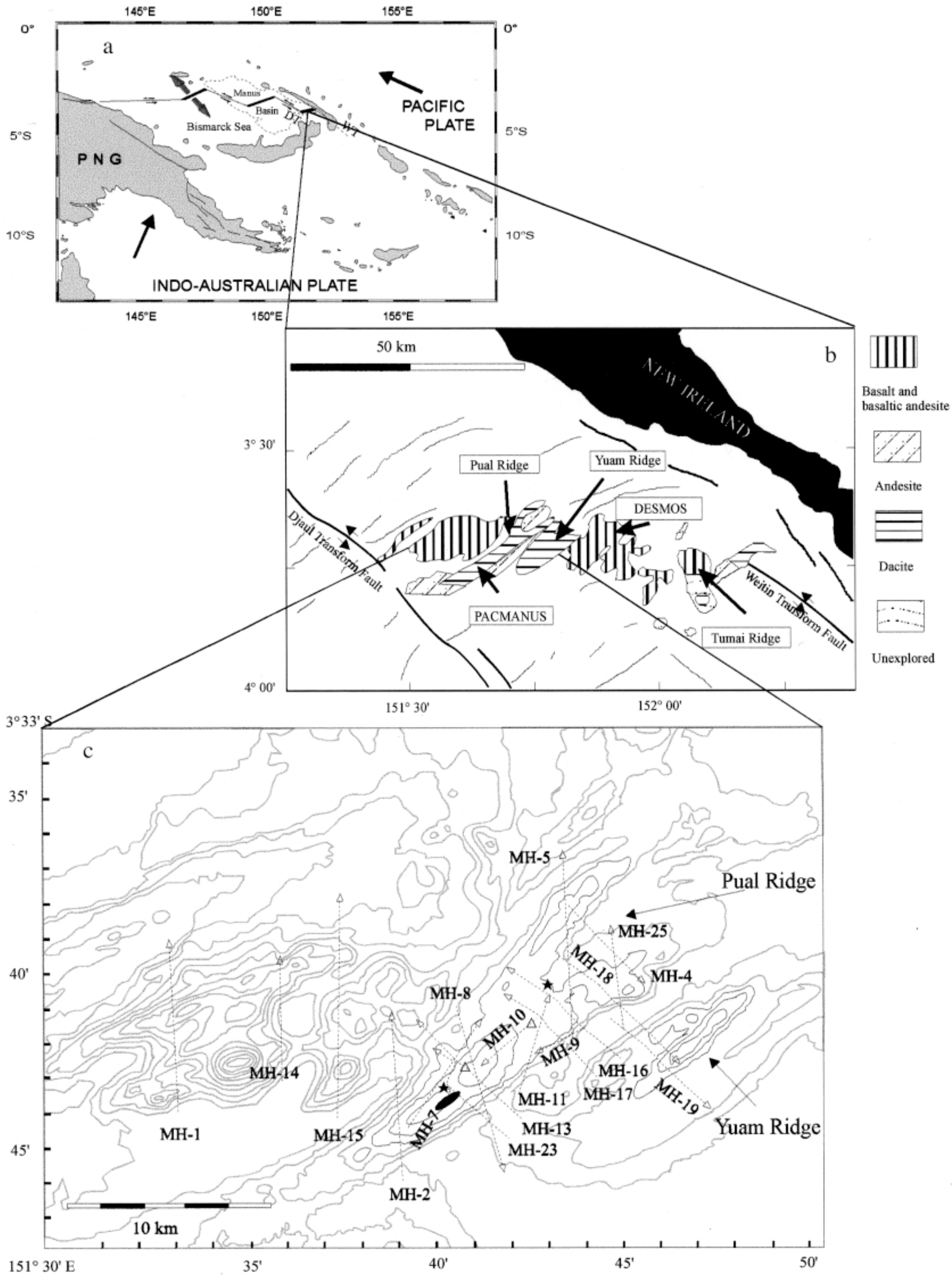
This study focuses primarily on the morphology, composition and size distribution of individual particles in the neutrally buoyant plume. We used a combination of gravimetric measurements and scanning electron microscopy coupled with X-ray energy-dispersion spectra (SEM–EDS), electron-microprobe analysis and image-analysis techniques.

Manus is the semi-enclosed back-arc basin of the volcanically active New Britain island arc. It lies geographically within the Bismarck Sea, located in the western equatorial region of the Pacific Ocean (Fig. 1a).

Eastern Manus is a pull-apart basin that lies between the Djaul and Weitin transform faults (Fig. 1a). The pull-apart structure is manifest as a series of *en échelon* volcanic ridges (Fig. 1b). This extensional structure is interpreted to be the result of oblique convergence of the northward-moving Indo-Australian plate and the northwestward-moving Pacific plate, against the New Britain trench to the south and the outer Melanesian arc to the east (Benes *et al.* 1994). According to Taylor (1979), the eastern Manus Basin was formed during the Pliocene (~3.5 Ma) by asymmetrical spreading of the seafloor at a fast rate, 13.2 cm/yr.

The neovolcanic zone within eastern Manus Basin is made up of several ridges, the most prominent of which is informally named Pual Ridge. It is approximately 20 km in length, trends NE–SW, and has a bathymetric minimum of about 1600 m (Fig. 1c). The northernmost portion of the ridge segment has two arms

FIG. 1. a. Regional tectonic setting of the eastern Manus Basin, Papua New Guinea, showing the two transform faults (Djaul, DT; Weiting, WT) that bound the extensional pull-apart basin. Arrows show relative motion of plates (modified from Benes *et al.* 1994). b. Map of the eastern Manus Basin showing general geology and locations of Pual Ridge, Desmos Cauldron and Tumai Ridge. The map also compiles the main composition of the area based on dredges (after Binns & Scott 1993, Scott & Binns 1995). c. SEABEAM bathymetric map (100 m isobaths) (H. Sakai, unpublished chart). The 1700–1900 m isobaths (solid lines) outline the Pual and Yuam Ridge segments. Dashed lines (at scale) represent CTD–T transects (all of them were within 10 km in length) studied during the PACMANUS expeditions in 1991 and 1993, and the arrows indicate the direction of the survey. The PACMANUS (black oval), other known hydrothermal deposits (Δ) (Binns & Scott 1993), and recently discovered sources (★) (Ortega-Osorio 1996) also are shown.


a
145°E
150°E
155°E
0°
5°S
10°S
Manus
Basin
Bismarck Sea
DT
WT
PNG
PACIFIC
PLATE
INDO-AUSTRALIAN PLATE
b
50 km
NEW IRELAND
3° 30'
4° 00'
151° 30'
152° 00'
Pual Ridge
Yuam Ridge
DESMOS
PACMANUS
Tumai Ridge
Djaul Transform Fault
Weitin Transform Fault
Basalt and
basaltic andesite
Andesite
Dacite
Unexplored
c
3°33' S
35'
40'
45'
151° 30' E
50'
10 km
MH-1
MH-2
MH-4
MH-5
MH-7
MH-8
MH-9
MH-10
MH-11
MH-13
MH-14
MH-15
MH-16
MH-17
MH-18
MH-19
MH-23
MH-25
Pual Ridge
Yuam Ridge

("Pual" means "fork" in a New Guinean dialect), which gradually deepen northeastward to 2000 m. Deep waters surround the ridge to the west and east, although another shorter ridge, "Yuam" in Figure 1c, lies nearby, to the east. The volcanic rocks that make up Pual Ridge are andesitic at the base and dacitic and rhyodacitic on top (Binns & Scott 1993, Scott & Binns 1995).

According to the latest descriptions of the Desmos Cauldron (Fig. 1b) reconfigured throughout multi-narrow-beam survey during a Japanese expedition (Gamo *et al.* 1997), the caldera is a 200-m-deep basin of about 2 km in diameter striking north–northwest. Highly acidic fluids of up to 120°C have been documented at this site of hydrothermal activity.

The Approach Used

An extensive survey (Fig. 1c) for hydrothermal plumes was conducted during the 1991 and 1993 PACMANUS expeditions in the eastern Manus Basin. Previous American, Japanese, and Russian cruises found indications in the water column of likely hydrothermal activity (Both *et al.* 1986, Craig & Poreda 1987, Tufar 1990, Scott *et al.* 1992, Binns & Scott 1993, Gamo *et al.* 1993, Lisitsyn *et al.* 1993). Figure 1c summarizes the operations during 1991 and 1993 expeditions [details of the CTD–T operations during the collection of suspended particulate matter (SPM) can be found in Ortega-Osorio (1996)]. The survey included along- and across-ridge-axis transects.

Particulate and thermal plumes, as indicators of active hydrothermal sources, were recognized and sampled by mean of a Conductivity – Temperature – Depth – Transmissivity (CTD–T) survey during both expeditions. This survey provided a real-time indicator of anomalous temperatures and concentrations of suspended particulate matter in the water column. By looking at the composition and distribution of the SPM, it was possible to infer the origin, proximity and maturity of the hydrothermal source.

The measurement of relative concentration of SPM is based on the decrease in light transmission. A Sea–Tech 0.25 m path-length transmissometer having an accuracy of ±0.5% was used for this purpose. The transmissometer's output was scaled from 0 to 100% for all stations. A General Oceanics CTD–Transmissivity rosette sampling system with 11 Niskin bottles and the transmissometer mounted on it was deployed and tow-yoed between 1500 and 50 m off the bottom along a predetermined track. The Neil Brown Mark III B CTD unit has a temperature accuracy of 0.005°C and a resolution of 0.0005°C, conductivity accuracy of 0.005 mmho and resolution of 0.001 mmho, and a 6400 dbar pressure transducer accuracy of 6.5 dbar and resolution of 0.1 dbar. Samples were taken where an evident hydrothermal signal was observed.

As soon as the device was retrieved from the sea, bottles were drained for various types of chemical analysis. Water samples were taken directly from the spigot of the Niskin bottles, which were occasionally shaken to maintain particles in suspension. The suspended particulate matter was retained with a 47-mm diameter and 0.40-μm pore size pre-weighed Nucleopore® polycarbonate membrane filter. Filtration was vacuum-assisted (~500–650 mm Hg) and normally took from 6 to 16 hours for small (4 L) and large (9 L) volumes, respectively. Care was taken to avoid breaking the filter membranes. After most of the water was filtered, bottles were rinsed with 100 mL of distilled deionized water and shaken in order to remove the remaining particles in the bottles as well as to wash out salt remaining on the filters. Filters were then removed with a pair of Teflon-coated tweezers from the filter holder in a clean fume hood, set in a sterilized plastic petri dish and desiccated for about five hours. Filters were finally freeze-dried under vacuum and stored frozen in an evacuated desiccator until analyzed onshore.

Analytical Methods

Morphological and chemical analysis of particles

Morphological characterization and elemental composition of individual grains in selected samples were determined by X-ray primary- and secondary-emission spectroscopy using a JEOL JSM–840 scanning electron microprobe (SEM) equipped with a PGT System 4 energy-dispersion X-ray spectroscope. Depending on the nature and composition of the particle, operating conditions were generally between 10 and 25 kV, working distance from 10 to 30 mm, probe current from 1×10^{-9} to 1×10^{-11} A, and magnification range from 100× to 30,000×.

Technique of sample preparation

Because contamination by foreign particles could occur during analytical procedures, special care was taken to minimize further manipulation after collection of the samples. A special 47-mm-diameter holder was designed for this purpose. This technique provided several advantages: i) the manipulation of the sample is restricted to the mounting procedure since no cutting or sectioning is involved; ii) a more representative analysis is obtained as the survey can be conducted on different portions of the filter (*i.e.,* during the filtration, particles are not spread homogeneously over the filter because of turbulent or concentric flow, resulting in a tendency for the material to concentrate randomly rather than only in the center of the filter or on an arc-segment), and iii) the sample is kept in storage for further analysis if needed, since this is a non-destructive technique. This special graphite-cored aluminum stub is sketched in Figure 2. The 47-mm aluminum disk supporting the filter was coated with carbon to avoid Al interference. Filters were first attached to the carbon-coated aluminum disk to keep them flat and to avoid curling caused by heat during the coating process. The filters were then sputtered with twice as much as the normal amount of graphite dust to obtain a conductive surface. This coating procedure resulted in a good conductive surface, with virtually no interference from aluminum, and is as effective as the palladium coating used in other studies (Feely *et al.* 1987, 1990, 1992). The filter and disk mounted on the aluminum stub were secured with an aluminum O-ring and placed into the SEM–EDS chamber for analysis.

Size distribution and relative abundance of particles

Image analysis was carried out on selected samples in order to obtain quantitative data of the size distribution and relative abundance of individual grains. The technique allows not only a measurement of the particle size in the sample, but also a distinction among different mineral grains, since chemical maps can be produced and combined. Images of Fe, Mn, S, Cu, Zn, Ca, Ba and Si X-ray maps were acquired by using a CAMECA™ electron-microprobe system. Operating conditions of the electron microprobe were optimized through several tests until satisfactory resolution was obtained (Ortega-Osorio 1996). Selected samples from close to (over the ridge) and far away from (~15 km) Pual Ridge were analyzed [the reader is refer to Ortega-Osorio (1996) for a complete description of the operating conditions of the electron microprobe]. The reproducibility of the technique was determined by scanning the same area several times. An apparent damage on the filter surface was noticed after the second and third scans, but despite this slight burning of the polycarbonate filter, the particles were not affected, and virtually identical images were obtained after three scans. Any variability in the results from different areas on the same sample thus stems from the nature of the sample itself and not from the instrument. Accuracy is difficult to evaluate (Russ 1990, 1992), since it depends greatly on the experience and preferences of the operator. For a given feature, the result can vary from user to user or even from time to time with the same user. Selecting the features of interest ("foreground") is an important prerequisite and requires that a "threshold operator" be defined. The threshold operator not only determines and separates the background from the foreground, but it also converts a gray-scale image (gsi) to a binary image, which occupies much less storage space in the computer. Furthermore, it is more suitable for image measurements (Russ 1990). As Russ (1990) has pointed out, the brightness histogram is probably the most useful tool for setting threshold levels of a given image. In this study, the threshold was defined at a fixed level using the brightness histogram of individual species and applied systematically throughout the analysis. Images of chemical maps were processed with the image analyzer Visilog™ software. The Visilog script program for analyzing plume-derived particles was presented by Ortega-Osorio (1996).

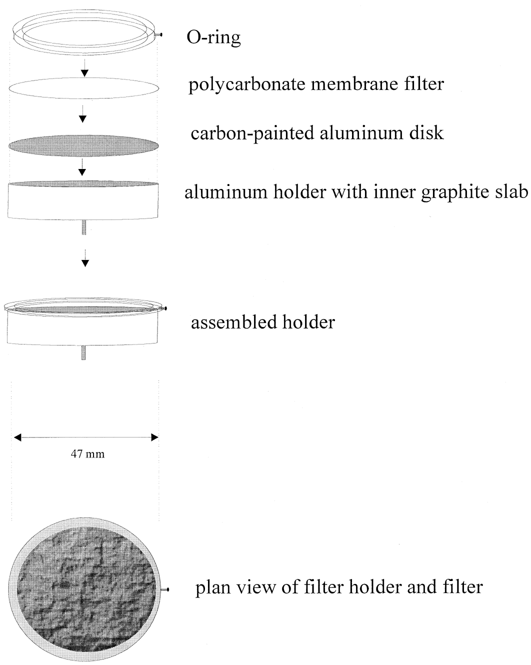


FIG. 2. Specially designed 47-mm filter holder for scanning electron microscope analysis of hydrothermal-plume-derived particulate matter.

RESULTS AND DISCUSSION

Recognition of plume particles

The CTD–T deep-tow surveys provided a means of estimating the extent and intensity of the hydrothermal discharge and led to the discovery of two new hydrothermally active sites (stars in Fig. 1c) at the south and north portions of the Pual Ridge (Ortega-Osorio 1996). These two sites will be referred to hereafter as the northern and southern plume sites. The measurements of SPM concentration within the non-buoyant hydrothermal plumes was compromised in some cases by our inability to distinguish unambiguously background samples from those with a hydrothermal component.

Relationship between SPM concentration and light-attenuation anomaly

Light transmission has commonly been used as an effective tracer of hydrothermal plumes. Its use provides accurate *in situ* measurements of beam transmission, which can, in turn, be related to the concentration of suspended particulate matter in the plume (Baker & Lavelle 1984). Although the attenuation of the beam does not depend entirely on the particle concentration, a linear relationship between these properties exists if other effects, such as particle size, shape and index of refraction, are negligible or mutually compensating (Baker & Lavelle 1984). Bartz *et al.* (1978) have pointed out that for a well-defined geographic region, the nature of the suspended particles does not change much and therefore, the optical technique is useful to determine total concentration.

A beam of light from a well-collimated monochromatic source can be attenuated by two basic processes, scattering and absorption. The energy removed by these processes is quantified as the light attenuation coefficient "c" (Bartz *et al.* 1978):

$$T = e^{-cr} \tag{1}$$

where T is measured transmission, r is length of optical path, and c is the attenuation coefficient (in units of m^{-1}). Therefore:

$$c = -\ln T / r \tag{2}$$

We assume that normal background conditions unaffected by hydrothermal activity prevail in the water column at some height above the plume. A systematic shift in transmission between single casts was removed by averaging the mean value of each dataset in order to normalize the background. A possible source of this deviation is a fogging deposition on the glass lenses of the transmissometer (Thomson *et al.* 1992b).

The relationship between SPM and Δc tends to be erratic and difficult to interpret. A plot of the total SPM against Δc has a low correlation ($r^2 = 0.2$). If values associated with a high error (*i.e.,* 5–10%) are excluded from the dataset, a weak linear relationship is obtained ($r \approx 0.5$). In general, however, light transmission was found not to be a reliable estimator of particulate concentration. Gravimetric determinations of plume-laden particles in the Juan de Fuca Ridge (Baker 1994) have shown an excellent agreement between SPM and Δc ($r^2 = 0.85$) after several years of sampling. Further surveys in the eastern Manus Basin in the future could improve our results.

Morphology and chemical composition of suspended particles

Early descriptions of sulfide precipitates around hydrothermal vents (Haymond & Kastner 1981) and in laboratory model experiments (Feely *et al.* 1987) have shown that the chemical composition of precipitates in the buoyant plume reflects the overall composition of high- and low-temperature sources at the mid-ocean ridges. PACMANUS chimney fragments are primarily composed of pyrite, chalcopyrite, anhydrite, bornite, tennantite, galena, sphalerite or wurtzite, silica and barite (Scott & Binns 1995). The fresh sulfide precipitates suspended in the rising plume might be expected to have this chemical imprint. However, measurements of particle fallout along the Juan de Fuca Ridge have shown that large (>30 μm) particles of sulfides (sphalerite, wurtzite, pyrite, pyrrhotite, barite, chalcopyrite, cubanite) settle within the first 10–20 m of dispersion (Converse *et al.* 1984, Feely *et al.* 1987), leaving those of smaller size (<2 μm) to be carried within the plume to greater distances. Moreover, Massoth *et al.* (1983), Nelsen *et al.* (1986), Feely *et al.* (1987, 1990, 1992), Walker & Baker, (1988), Trocine & Trefry (1988) and German *et al.* (1990) have suggested that oxidative dissolution and scavenging are responsible for depletion of metal sulfides as the plume rises and disperses horizontally, such that the particles apparently lose most of their sulfide component as they travel away from the source.

Suspended particulate matter was examined in detail on filters with the SEM–EDS approach. Preliminary observations at low magnification (500–1000×) revealed abundant homogeneously distributed small ($\lesssim$20 μm) particles. In Back-Scatter Emission mode, we identified particles of high atomic number (*i.e.,* iron, silicates, barite, sulfides). Higher magnification (10,000–50,000×) revealed the main morphological features of these particles. Replicate point and bulk analyses of individual particles were done. The predominant particles in the eastern Manus Basin plumes are anhydrite, barite, an unidentified Si–Fe-rich phase, colloidal amorphous iron oxyhydroxides, Fe–Mn oxyhydroxides of apparent bacterial origin, and a very diverse background array of biogenic detritus (shrimps, diatoms, silicoflagellates, copepods and unidentified gelatinous organic matter).

The SEM examinations did not allow us to determine the internal ultrastructure of apparent bacterial matter; they only provided images of the external morphology and elemental composition (Cowen 1991). However, observations of similar hydrothermal particulate matter from the Juan de Fuca Ridge by transmission electron microscopy have revealed internal structures made up of bacteria encapsulated by an Fe–Mn phase in an extracellular matrix of organic polymers (Cowen *et al.* 1986).

Seven major groups of particles have been identified on the basis of their morphology and composition: i) filamentous particles of Mn–Fe–Si–Cu–Zn with minor Mg–Al–P–K–Ca, ii) orb-like particles of Fe–Mn–Si with minor Mg–Al–P–K–Ca, iii) anhedral barite, iv) euhedral anhydrite, v) amorphous iron-rich phase, vi) an unidentified Si–Fe rich phase, and vii) biogenic material. The elements are listed in order of their decreasing relative abundance based on peak intensities in the X-ray spectra.

Survey of the Seven Groups of Particles

Filamentous particles rich in Mn–Fe–Si–Cu–Zn with minor Mg–Al–P–K–Ca

These particles vary in length from ~10 to 30 μm and are generally characterized by their "filamentous" structure (Fig. 3a). This group of particles is common in most samples, but the composition varies from site to site. There is a distinct enrichment of Cu and Zn in samples above the southern site at the PACMANUS deposit, but not at the northern site. Although bacterially mediated precipitation is considered to be the controlling process of metal concentration, Cu and Zn enrichment might depend on their availability rather than bacterial affinity for chalcophile elements. The presence of these elements suggests proximity to a source, since chalcopyrite and sphalerite are dense phases that precipitate and fall out shortly after the vent fluid is ejected (Feely *et al.* 1987, 1990). The higher peak for Cu relative to Zn is consistent with the dominant mineralogy in chimney fragments from the PACMANUS deposit (Scott & Binns 1995). Cu-rich particles were not detected at the northern site. The enrichment in both Fe and Mn is not surprising, since some microbes at hydrothermal vents accumulate both elements on their surface (Juniper & Tebo 1995). Laboratory studies of hydrothermal mesophilic manganese-oxidizing bacteria (Ehrlich 1983, 1985) demonstrate that some of the most important enzymatic reactions between bacteria and end-member vent solutions involve the oxidation of Mn^{2+} and Fe^{2+}. Nevertheless, there is still a gap in our understanding of the mode of existence of these microbes and of their relationship to metal-rich hydrothermal fluid because isolation of the microbes is difficult (Ehrlich 1983, 1985). Whatever the ecology or physiology of the microbes might be, scavenging reactions are considered to play a significant role in both concentration and distribution of particulate and dissolved manganese in the ocean (Cowen *et al.* 1986). Besides, a number of mechanisms, such as self-oxidation and enzymatic reactions, has been suggested for the precipitation of manganese, and its coprecipitation with iron oxides (Edmond *et al.* 1982, Weiss 1977). Particulate matter of fallout origin at plume sites is now well recognized as the main mechanism of dispersion and concentration of manganese into metalliferous sediments (Weiss 1977). However, the inventory of manganese oxide deposits is still limited and probably underestimated.

Aggregates of Cu-rich sulfide were observed from the plumes above the Desmos Cauldron. This type of particle is virtually absent from all other sites.

Orb-like particles rich in Fe–Mn–Si with minor Mg–Al–P–K–Ca

"Orb-like" particles have a distinct spherical shape and a wool-like texture (Fig. 3b). They range from 2 to 5 μm in diameter and, as is the case for the filament-like group, they are generally observed in all samples, although they are less abundant. A noticeable difference between these two groups is that orb-like particles tend to have high Fe:Mn values and have no traces of copper and zinc.

Spontaneous oxidation of reduced iron occurs rapidly as the hydrothermal fluid interacts with normal seawater, resulting in a common iron oxyhydroxide phase (Lupton *et al.* 1998, Bowers *et al.* 1985, Stumm & Morgan 1981). However, microbially mediated precipitation has been documented to be equally important (Cowen *et al.* 1986, 1990).

Hydrothermal-symbiotic colonies of bacteria around hydrothermal vents have been reported from a variety of environments such as the Juan de Fuca Ridge (Tunnicliffe & Fontaine 1987), 21°N EPR (Alt 1986, 1988), and Galapagos Rift (Ehrlich 1983). Conversely, there is limited knowledge about whether or not the bacteria are specific to hydrothermal environments. Chemo-autotrophic bacteria can be carried by advection within hydrothermal plumes, or they may already be in the water column and simply take advantage of nutrients and metal-rich waters (Juniper & Tebo 1995). Among the large variety of iron-oxidizing bacteria, *Gallionella sp.* and *Leptothrix sp.* are probably the most common reported in the vents. Spherical, bacillus and filamentous structures are typical morphological features. Spheres of similar size to the orb-like particles from eastern Manus Basin are known from the Juan de Fuca Ridge (Juniper & Tebo 1995). Because of the presence of discernible sheaths of bacteria within the spheres, Juniper & Tebo (1995) suggested that aggregates of bacteria are necessary to promote the formation of iron-rich spheres. Tunnicliffe & Fontaine (1987) concluded that iron first accumulates as fine submicrometric particles around and between the sheaths. Both observations indicate that bacterially mediated precipitation requires a substrate, and this continuous process of aggregation and cementation eventually leads to a "solid" sphere. At the eastern Manus Basin, this phenomenon was observed at the northern plume site, where different stages of formation of iron-oxidizing bacteria were observed overgrowing platelets and spheres of a Si–Al-rich phase. The apparent incipient stage of formation of these particles indicates that early chemical evolution within the plume is still in progress and, therefore, the particles are relative young. A recently activated source at this northern site (Ortega-Osorio 1996) could explain this situation.

Another morphological group of iron-oxidizing bacteria was found at the northern plume site. These have a filamentous structure similar to filament-like particles of high Mn:Fe ratio (probably manganese-oxidizing bacteria) found in the same area. From morphological and compositional considerations, they appear to result from two distinct bacterially mediated formations: (1) the iron-oxidizing and (2) the manganese-oxidizing bac-

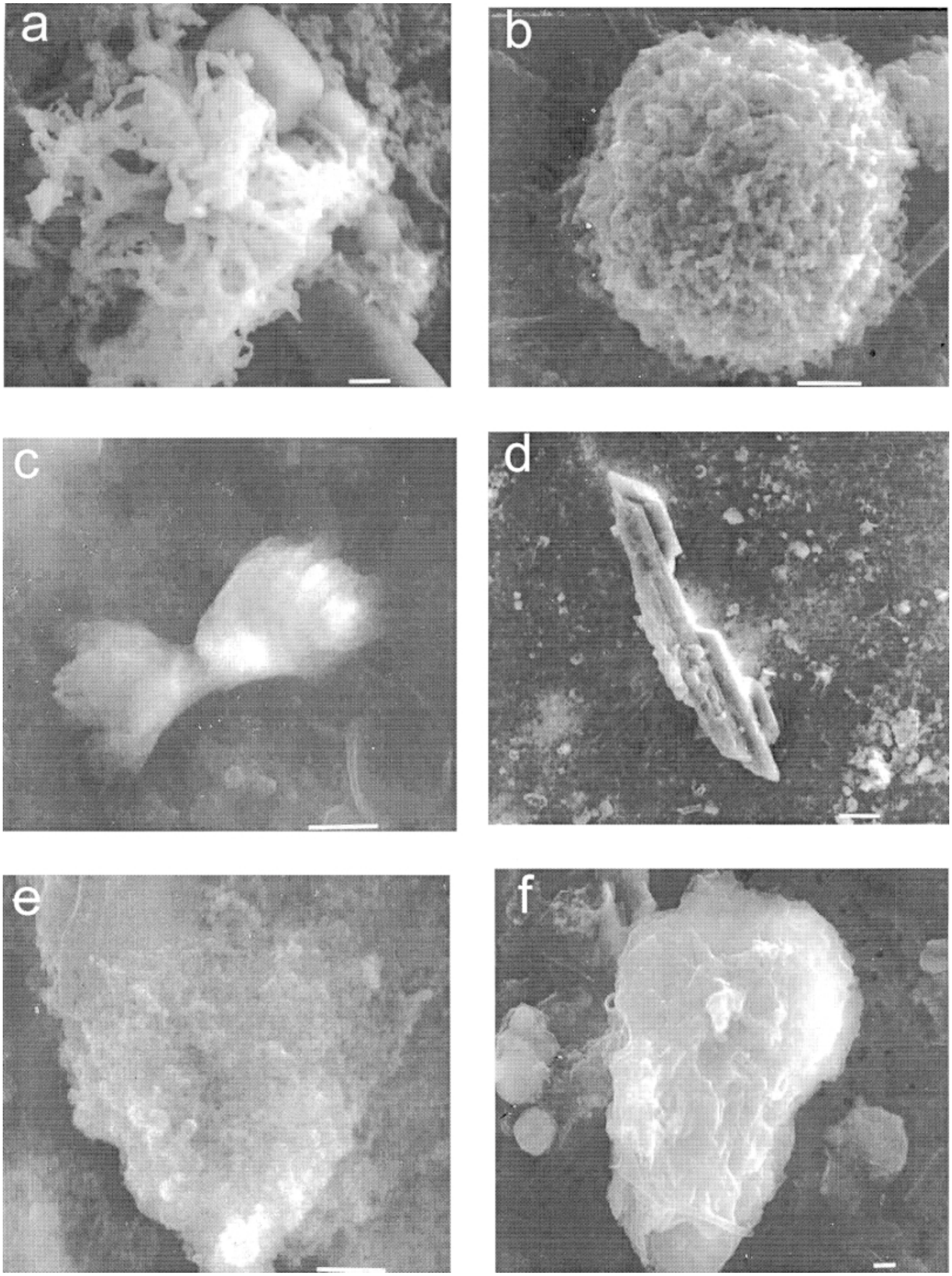


FIG. 3. Scanning electron micrographs and EDX spectra of neutrally buoyant plume-derived particles from the eastern Manus Basin. The collection of spectra in all cases was carried out for a period of 100 seconds and an accelerating voltage of 15–20 kV. (a) All particles in this group exhibit typical manganese-oxidizing bacterial filaments. Mn–Fe-rich particle with Cu and Zn contents; this sample was collected above the southern active source on survey line MH–7. (b) Particles in this group

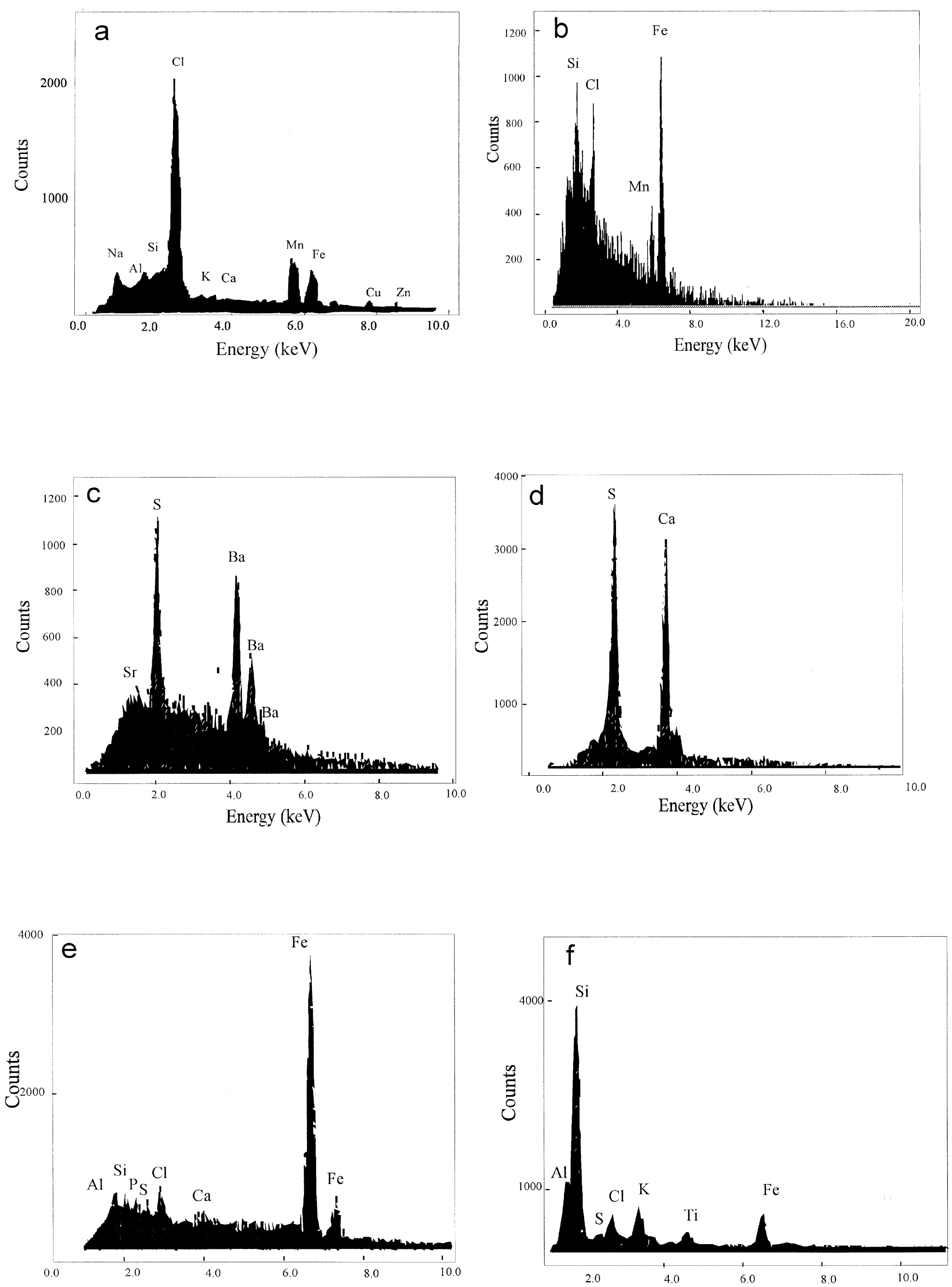


exhibit typical iron-oxidizing bacterial spheres and filaments. The Fe–Mn-rich sphere was collected above the southern active source on survey line MH–7. (c) Anhedral particle of barite collected at the northern active source on survey line MH–5. (d) Particles of anhydrite taken above the southern active source on survey line MH–7. (e) Submicrometric amorphous iron with organic particle collected above the southern active source on survey line MH–7. (f) Si–Fe-rich particles taken above the southern active source on survey line MH–7. The scale bar in all micrographs is 1 μm except in (d), where it is 10 μm.

teria. Lundgren & Dean (1979) have determined that the *Leptothrix–Sphaerotilus* group of bacteria precipitates filamentous sheaths encrusted with hydrated ferric and manganiferous oxides. Iron filament formations seem to be most common around iron oxyhydroxides deposits. Occurrences of filamentous bacterial mats at Franklin Seamount in western Woodlark Basin, Papua New Guinea (Binns *et al.* 1993) and southern Pacific hot spots and the EPR regions (Hekinian *et al.* 1993) have been well documented.

Barite particles

Particles of barite typically have an anhedral morphology and are generally submicrometric to ~4 μm in size (Fig. 3c). This group of particles occurs rarely at some sites. Grains have an overgrown appearance. Barite is one of the main constituents of the hydrothermal precipitates, resulting from the end-member solution – seawater interaction (Trocine & Trefry 1988). Together with large particles of sulfide phases, these particles settle quickly from the rising plume. Occurrence of barite in the non-buoyant plume from the northern source and from the Desmos Cauldron suggest that these particles could have been formed recently.

Anhydrite particles

Particles of anhydrite were found at both the southern and northern plumes sites above the Pual Ridge. Grains are generally larger than the other suspended particles. They form irregular euhedral shapes with rounded edges and range in length from 20 to 50 μm (Fig. 3d). They are abundant in most samples, but not in background samples. Their shapes include elongate tabular particles to irregular but still angular particles. Larger particles were observed at the northern source (Fig. 1c) and from the Tumai Ridge (Fig. 1b). Particles from the Desmos Cauldron (Fig. 1b) exhibits a morphology typical of rapid growth.

Field observations and laboratory experiments (Feely *et al.* 1987) have shown that the dissolution of anhydrite is rapid. This mineral thus is a very reliable indicator of recent local hydrothermal activity where it is found within a neutrally buoyant plume.

Amorphous iron-bearing phase with minor P–Mg–Al–K–Ca–Mn

This group is characterized by a distinctive submicrometric size and amorphous structure. It is present in the entire area, above the Pual Ridge (Fig. 3e), at Desmos Cauldron and Tumai Ridge. Some particles are accompanied by organic aggregates with recognizable features, such as specula or spines that probably work as a substrate for initial aggregation, as was described for the iron oxyhydroxides. Amorphous iron-bearing material has been reported as a typical hydrothermal precipitate from several areas (Dymond & Roth 1988, Trocine & Trefry 1988, Feely *et al.* 1990, 1994b), and it is usually accompanied by bacterially mediated iron oxyhydroxides (Cowen *et al.* 1986).

Unidentified Si–Fe-rich particles with minor S–Ca–K–Al–Mg–Ti

This group of particles has a variable composition, is generally irregular in shape, is abundant, and consists mainly of large grains up to 100 μm across (Fig. 3f). On the basis of descriptions of a similar phase from the Juan de Fuca Ridge (Feely *et al.* 1987, Von Damm & Bischoff 1987), these particles probably consist of ferroan talc stable at hydrothermal vent conditions.

Organic particles

Abundant organic particles, many of them recognized organisms, are observed in some samples. Size and shape vary from amorphous clusters to well-preserved silica-rich skeletal disks to Fe-rich membranous sheets (Al–Si–P–S–Ca) of unidentified organic matter. Diatoms and silicoflagellates are identified as a part of this group (Barron 1993, Lipps & McCartney 1993).

A high concentration of biomass has been reported within deep hydrothermal plumes at the Juan de Fuca Ridge (Winn *et al.* 1986, Thomson *et al.* 1992a). We measured the highest anomalies in transmission of light (highest concentration of particulate matter) at the Desmos Cauldron, a site of known active hydrothermal venting (Gamo *et al.* 1993). Large amounts of unidentified organic matter were found at this site, which suggests that the plumes may promote enrichments in organic matter. Al concentrations in the EDX spectra indicate that resuspension of bottom sediments also might occur. On the basis of its elemental signature (Mg, K, S, Cl, P, Ca), Dixon *et al.* (1995) claimed that similar matter from the TAG area was protoplasm. This unidentified organic matter likely represents most of the total SPM concentration determined at Desmos.

Size distribution and relative abundance

Information on particle size is important in evaluating chemical changes (dissolution and scavenging) during dispersal and settling processes that affect plume-derived suspended matter. Figure 4 illustrates three representative histograms of size distribution measured in this study; Table 1 summarizes the most relevant results.

The size distribution in the histograms is expressed in units of area (μm^2). The relative abundance of individual particles has been estimated from their total number. More than 50% of all particles fall within the range of ~2 μm in diameter. Larger particles are more randomly distributed, and they never dominate the distribution. However, on a mass basis, a single large particle can be equivalent to many small ones. Particles from

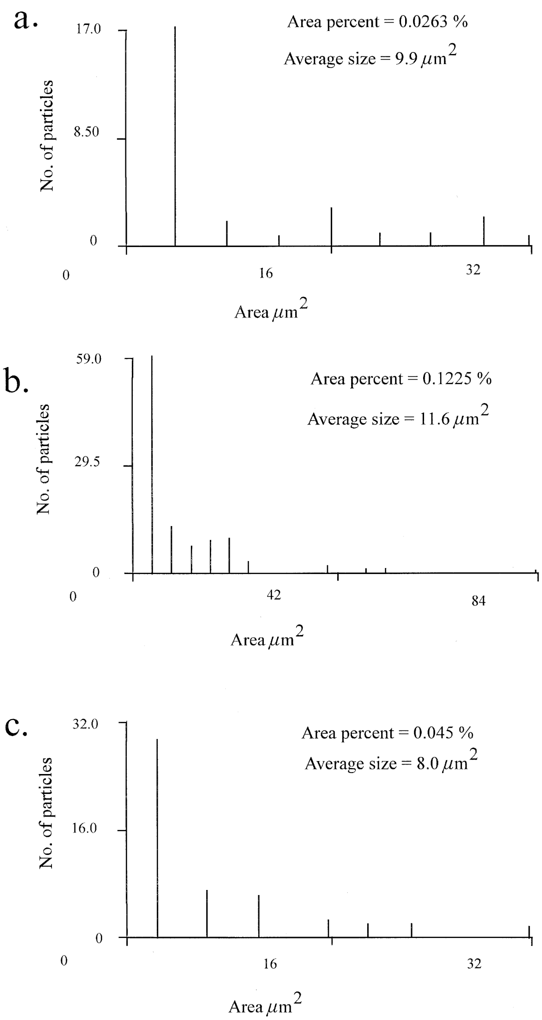


FIG. 4. Histograms of size distribution of neutrally buoyant plume-derived particles at the eastern Manus Basin. (a) Sample collected at survey line MH–1. (b) Sample collected above the southern active source on survey line MH–7. (c) Sample collected above the northern active source on survey line MH–16.

groups I and II (filamentous and orb-like) show this compensating effect between the southern and northern sites. The former consists of larger particles (~12 μm) lower in abundance (4.5×10^4), whereas the latter contains particles of smaller size (~8 μm) but of greater abundance (7.3×10^4) (Table 1).

Because manganese has a slower rate of oxidation in seawater than iron, bacterial oxidation is considered to be an important factor in removing dissolved manganese from seawater, in which case the abundance of particulate manganese should increase with time (Cowen *et al.* 1986, Cowen 1991). Therefore, it can be inferred from the Mn:Fe ratio in Table 1 that the plume at the southern site is more mature than the one at the northern site. Also, particles of anhydrite at the northern site are of larger size and are more abundant, which indicates a younger stage of dissolution in comparison to the southern site.

Hydrothermal plumes over the Pual Ridge have a higher concentration of particulate manganese than elsewhere in eastern Manus Basin. The relatively low Mn:Fe values at Desmos Cauldron and Tumai Ridge not only indicate that iron oxyhydroxides are a dominant phase, but also that these sites contain immature plumes. Large and abundant particles of anhydrite in both regions support this observation.

Neutrally buoyant hydrothermal plumes are characterized by high concentrations (150–350 μg/L) of suspended matter in the core of the plume. SPM concentrations decrease toward the outside limits of the plume to background values (20 μg/L). The weak linear relationship between amount of SPM and light-attenuation anomaly (Δc) arose from insufficient washing of the filters, which led to unacceptable contamination by sea salt. Consequently, it was not possible to map concentration of particulate matter within the plume by transmissometry.

The coexistence of different groups of particles (I and II) in the same sample could be interpreted as preferential bacterially mediated precipitation of manganese or iron (or both). Whether or not the bacteria are specific for the hydrothermal environment remains unknown, but we suggest two main mechanisms to explain their presence within the plume: 1) resuspension of existing bacterial mats around the source followed by advection within the rising plume and dispersion, and 2) presence of "normal" oceanic bacteria that multiply because of nutrients in the hydrothermal metal-rich plume.

Because chemical reactions and dissolution processes are imprinted on the particles, the composition of suspended hydrothermal matter could reflect the age of the plume. The bacterial scavenging of dissolved manganese results in an increase in the distribution of manganese-bearing particles with distance from the source and provides a rough to estimate of the relative age of the plume (Cowen *et al.* 1990). Suspended matter within the plume over Pual Ridge has a composition different from that at Desmos Cauldron and Tumai Ridge. Particles in the neutrally buoyant plume consist primarily of <2 μm anhedral iron oxides and 100 μm anhydrite and Si–Fe-rich phases.

Although size distributions of hydrothermal particles in the eastern Manus Basin are characterized by high concentrations of small particles up to 2 μm, large particles also are observed close to active sources. It is important to point out that these particles do not dominate the distribution, however, probably because they

TABLE 1. PARTICLE GROUP, DOMINANT MORPHOLOGY, AVERAGE SIZE AND RELATIVE ABUNDANCE OF PLUME-DERIVED PARTICLES OF REPRESENTATIVE SAMPLES FROM VARIOUS LOCATIONS IN THE EASTERN MANUS BASIN HYDROTHERMAL FIELD

				Group I and II filaments, spheres			Group III anhedral			Group IV euhedral			Group V amorphous			Group VII aggregate
	Depth m	SPM μg/L	Mn/Fe	Size μm	No. particles	Ab.	Size μm	No. particles	Ab.	Size μm	No. particles	Ab.	Size μm	No. particles	Ab.	Ab.
Background sample MH-1/5	1580	1245	0.97	9	4.5	xx			-			-	4	54	xx	x
Southern plume MH-7/5	1670	241.1	1.75	12	4.4	xx			-	5.2	0.98	x	23	860	xxx	xx
Northern plume MH-16/9	1641	467.8	1.47	8	7.3	xx	4	14	x	31	3.5	xx	10	120	xx	xx
Desmos Cauldron MH-27/5	1641	152.7	0.20	4	0.6	x			-	50	21	xx	56	710	xxx	xxx
Tumai Ridge MH-28/1	1463	449.7	0.14			-			-	43	78	xxx	12	120	xx	xxx

Groups: I filament-like particles, II orb-like particles, III barite, IV anhydrite, V iron-rich particles, VI Si–Fe rich particles, VII particles of organic matter. Size: mean values are given. No. particles: the number of particles ($\times 10^4$) is calculated from the number of particles observed in a ~1 mm^2 field times the effective area of filtration (14 cm^2). Ab.: estimated relative abundance. xxx: abundant, xx: moderate, x: scarce, -: absent.
Group VI was not included in this analysis. Because of the chemical and physical nature of the material of particles of group VII, only the Ab. is shown.

settle faster and because they could be dissolved faster in the water column. The finest hydrothermal particles are capable of being dispersed over a wide area and eventually fall out of the plume, contributing a component to the underlying metalliferous sediments in the basin, as has been inferred from geochemical studies of the underlying sediments (Ortega-Osorio & Scott 1994).

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