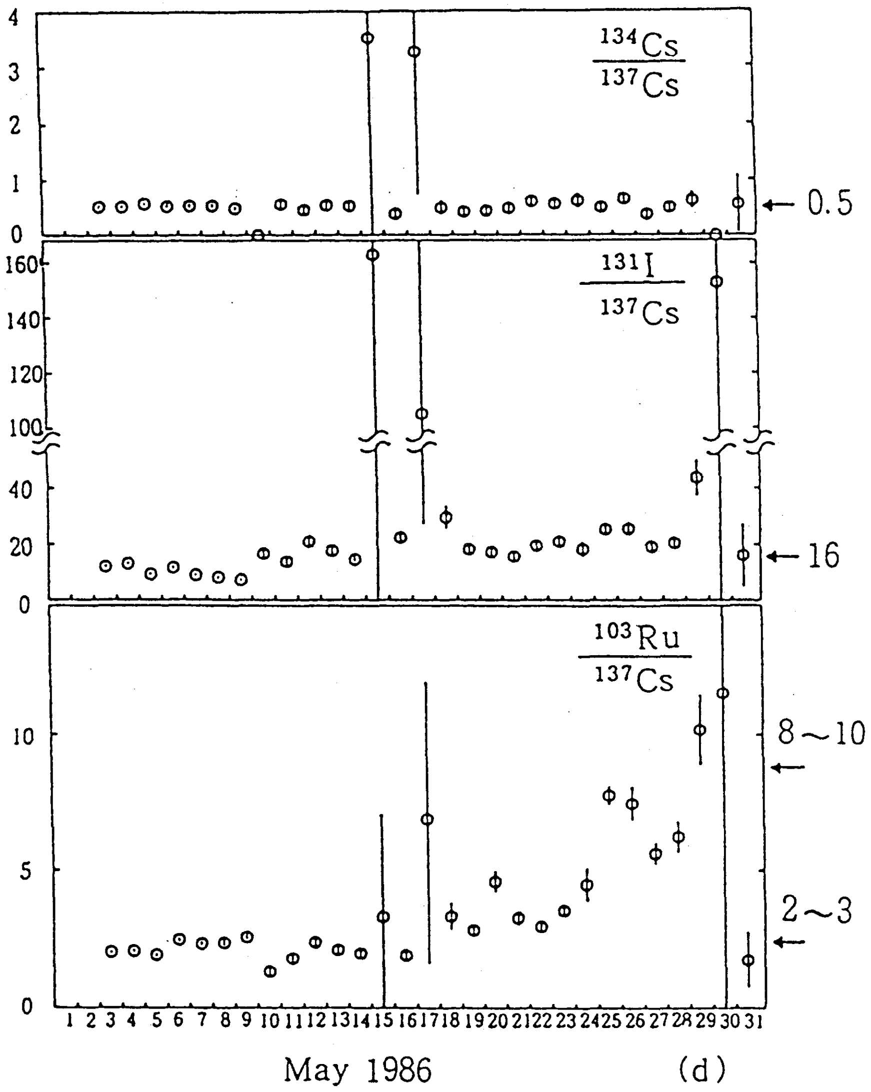

Fig. 96-23 Temporal variation of activity ratios of Chernobyl-derived radionuclides in surface air at Tsukuba.

ORGANIC MATTER AND LIGANDS

5. Ocean Biogeochemistry

5.1 Characterization of particulate protein in Pacific surface waters

Tanoue (1996)

Dissolved organic matter (DOM) is one of the largest but most poorly understood active reservoirs of organic matter on the planet. Although most DOM is marine in origin, its sources and sinks are not well known. As referenced in the 1995 ACTIVITIES, Tanoue extracted dissolved proteins from seawater and found that a limited number of protein species accounted for most of the dissolved proteins and that proteins contributed quantitatively to dissolved organic N throughout the water column.

Protein is the major cellular constituent of phytoplankton and 85% of phytoplankton nitrogen is in the form of protein (e.g., Billen, 1984). Cellular proteins in living organisms may be converted to detrital "combined amino acids" or "proteinaceous compounds" through biogeochemical processes. Particulate-combined amino acids (PCAA), the largest identified fraction of particulate organic matter (POM) in oceanic surface waters, represent a mixture of cellular proteins of organisms and of detrital combined amino acids. The dynamics of the two types of PCAA are expected to be quite different, but have not been distinguished in previous PCAA studies.

Tanoue (1996) reported the molecular characteristics of particulate proteins in surface waters along transects from 45°N to 25°S in the central Pacific (Fig. 96-24). The majority of PCAA was in the form of protein molecules in samples from the northern Pacific and Equatorial regions, namely, productive areas, while PCAA was mainly present as nonproteinaceous amino acid in subtropical regions, namely, oligotrophic areas (Fig. 96-25). Thus, it appears that the chemical form of PCAA, one of the major constituents of POM, varies meridionally.

Two typical groups of particulate protein were identified from meridional differences in molecular distribution (Fig. 96-26). The first group, derived directly from cellular proteins of living organisms, was made up of a large number of proteins, each present at a relatively low level, which gave smeared electrophoretograms

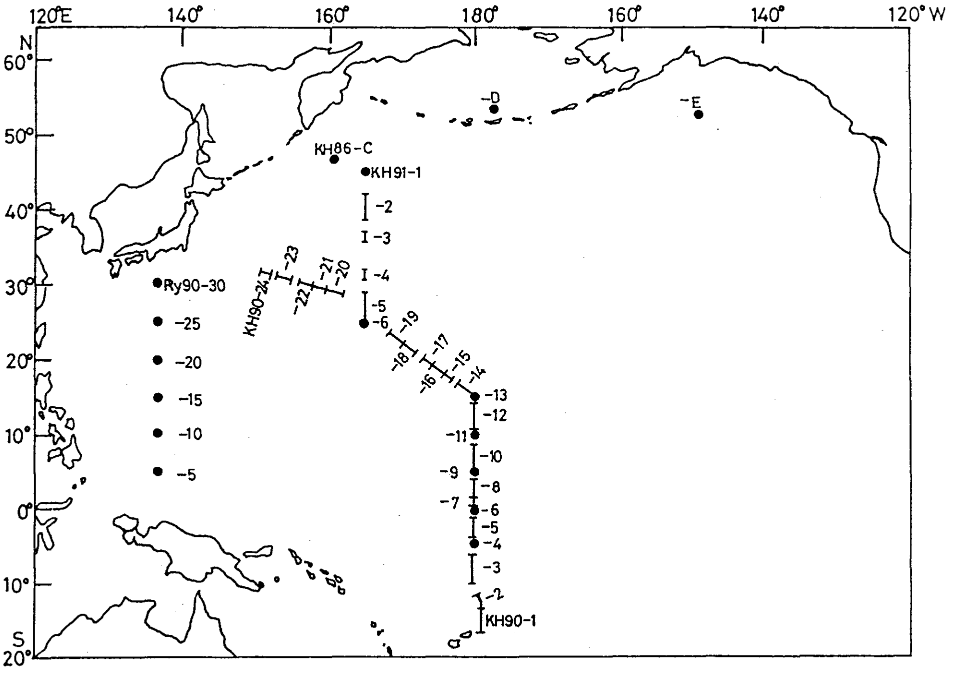

Fig. 96-24 Locations at which particulate matter in surface waters was sampled during cruises Ry90-01 (sample nos. Ry90-5 to Ry90-30), KH90-2 (KH90-1 to KH90-24), and KH91-3 (KH91-1 to KH91-6). Stations (KH86-C, -D and -E) in northern North Pacific and in Bering Sea for which SDS-PAGE patterns of particulate proteins were previously reported (KH86-D and -E; Tanoue, 1992) are also shown.

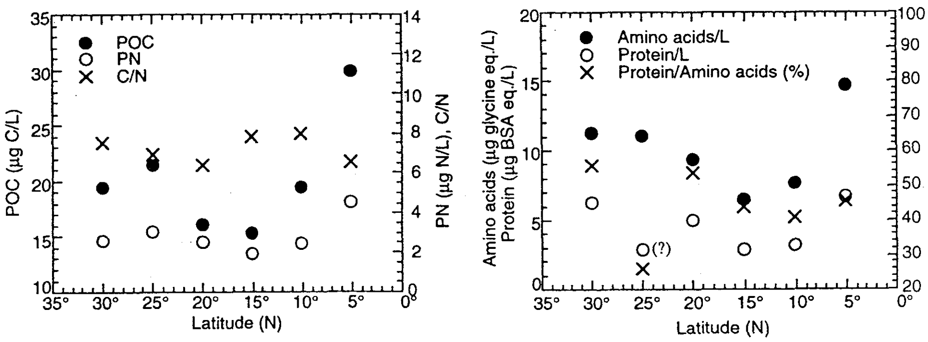

Fig. 96-25 Concentrations of POC (μg C/l), PN (μg N/l), PCAA (μg relative to a glycine standard/l) and particulate proteins (μg relative to a BSA standard/l), and C/N and Protein/PCAA (%) along 137°E (Ry90-01).

and were considered to be "background" proteins that contributed greatly to both total protein and PCAA, and appeared to be readily remineralized. The second group included a small number of specific proteins with a limited range of molecular mass. This group was prevalent in oligotrophic areas, an indication that proteins from specific sources survive and accumulate due to their resistance to degradation.

A protein with an apparent molecular mass of 45 kilodaltons (kDa), a member of the second group, was commonly found at low latitudes and the partial N-terminal amino acid sequence indicated that the 45 kDa protein was a single protein species that has not previously been reported (Table 96-3). Thus, a single identifiable protein molecule appears to be very widespread at low latitudes.

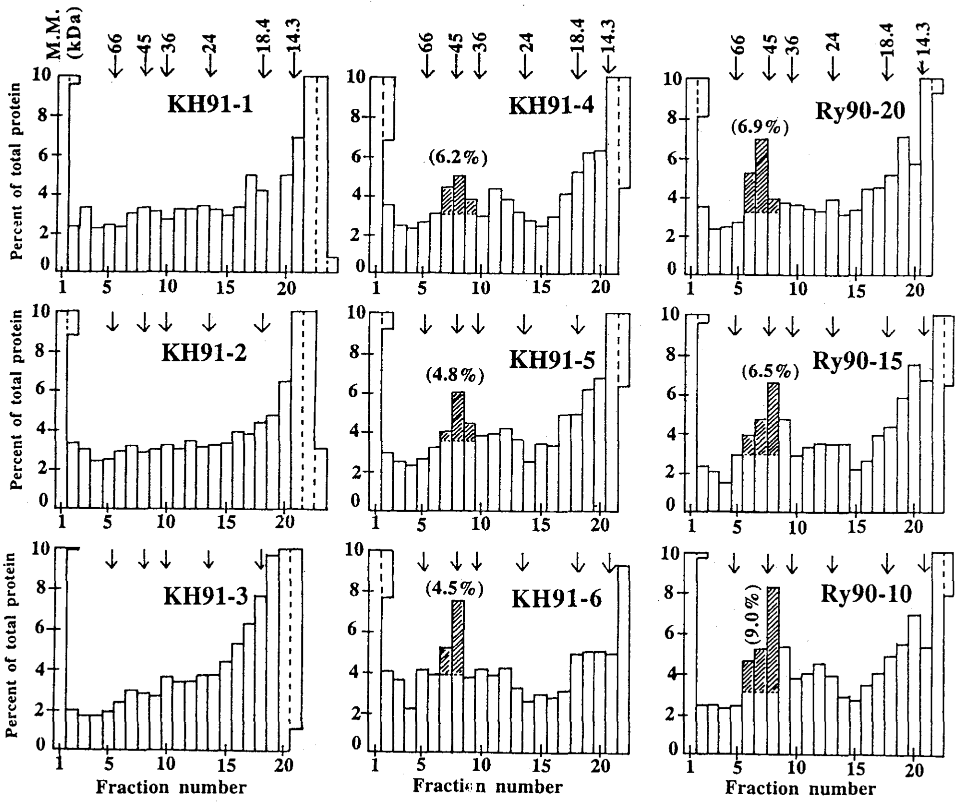

Fig. 96-26 Relative abundance of proteins in 5-mm pieces of gel for samples collected along 165°E and 137°E. Shaded fractions represent contribution (%) of proteins in distinct bands to total protein.

Table 96-3 N-terminal amino acid sequence of the 45-kDa protein obtained by automated Edman degradation.

Cycle	1	2	3	4	5	6	7	8	9	10	11	12
45-kDa protein	Gly	Thr	Gln	Pro	Asn	Pro	Ser	Pro	Ala	Ser	Pro	Val

5.2 Discrete dissolved and particulate proteins in oceanic waters

Tanoue, Ishii, and Midorikawa (1996)

Tanoue *et al.* (1996) extracted dissolved and particulate proteins from samples of surface seawater collected from the equatorial area, through the Indian Ocean, to the Antarctic Ocean (Fig. 96-27). Dissolved proteins were also observed in waters of the equatorial Pacific. They detected dissolved and particulate proteins with a wide range of molecular masses by sodium dodecylsulfate-polyacrylamide gel electrophoresis (SDS-PAGE). The particulate proteins were made up of many background proteins of overlapping molecular weight, which caused uniform staining in gel. However, distinct bands of individual proteins with apparent molecular masses of ~66 and 45 kDa were evident among background proteins (Fig. 96-28).

Electrophoretograms of dissolved proteins were quite different from those of the particulate proteins. Dissolved background proteins were not significant, and fewer than 30 proteins were clearly visualized as major dissolved proteins. Dissolved proteins with apparent molecular masses of 48 and 37 kDa were commonly found as major proteins in all samples examined.

Such molecular characteristics of dissolved and particulate proteins are consistent with previous results from the North Pacific. It thus appears that processes by which specific proteins from marine organisms are transferred to and accumulated in pools of dissolved and particulate organic matter are identical throughout the world's oceans.

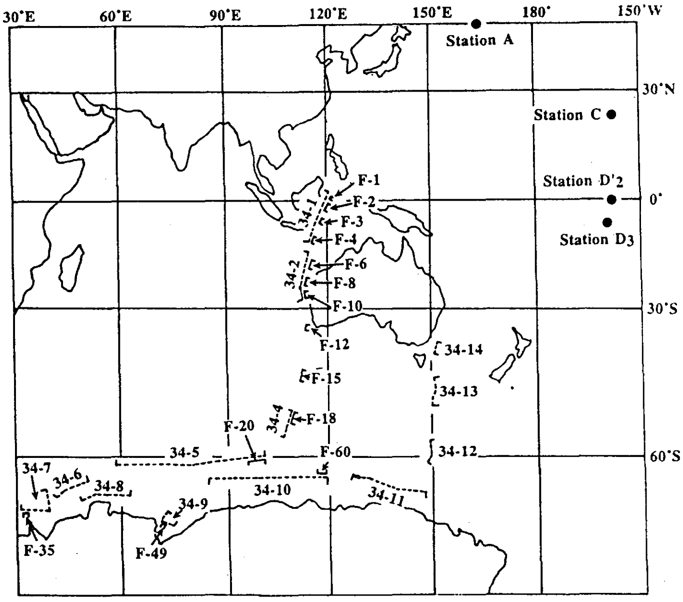

Fig. 96-27 Distribution of sampling locations of composite samples of surface seawater for extraction of dissolved proteins (broken lines: sample 34-1 to 34-14) and samples of surface POM for particulate proteins (solid lines: F-1 to F-60) along the track of JARE 34, and locations of depth stations A, C, D'_2, and D_3 for extraction of dissolved proteins during cruise KH 93-4.

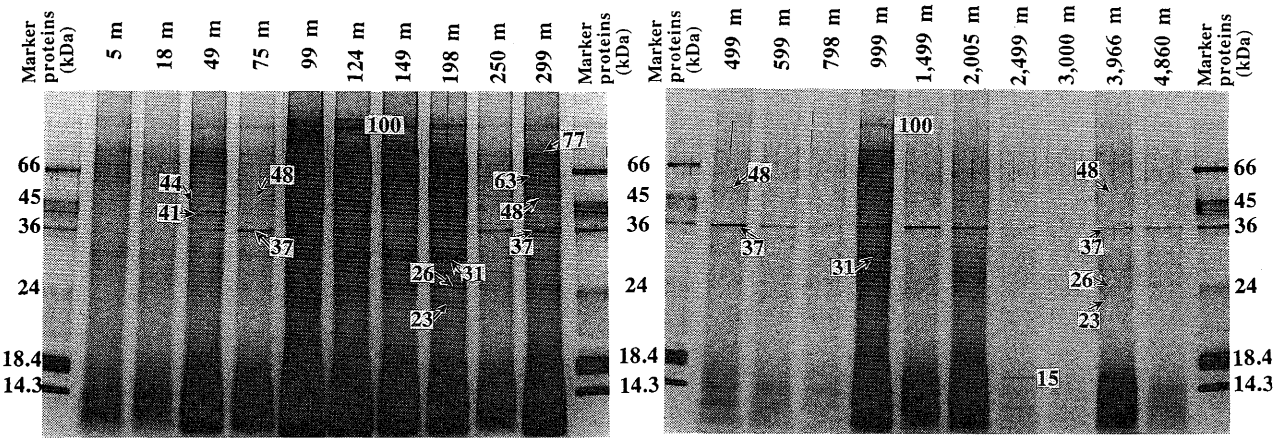

Fig. 96-28 Depth profiles of dissolved proteins at station D′₂. Proteins were, visualized by silver staining. Amounts of sample loaded on gel were equivalent to 100 ml of original seawater in each case. Each marker protein was loaded at 25 ng in each left- and 50 ng in each right-hand lane of each gel. Electrophoretograms of samples from depths of 18, 2005, and 3996 m were reported elsewhere (Tanoue 1995).

5.3 Abundance of viruses in deep oceanic waters

Hara, Koike, Terauchi, Kamiya, and Tanoue (1996)

Viruses are recognized as important members of the marine surface water ecosystem due to their role as decomposers of bacterial and protistan biomasses. The concentration of viruses is information essential to the ecological study of the aquatic environment.

Hara *et al.* (1996) investigated vertical distributions of bacteria and viruses at oceanic stations located in subarctic (Stn A) and subtropical (Stn B) areas of the Pacific using direct count and transmission electron microscopy (Table 96-4). Small DAPI-positive, virus-like particles (VLP) were found to be distributed throughout the water column down to 5000 m depth at both stations. The abundance of VLP ranged from 38×10^5 ml^{-1} at 50 m depth to 0.6×10^5 ml^{-1} at 5000 m depth at Stn A (Fig. 96-29). The ratio of VLP to bacteria-like particle (BLP) ranged from 1.1 to 7.4 at Stn A and 1.0 to 8.7 at Stn B in the entire water column. The maximum ratio was recorded at Stn B from the deepest sample, collected at a depth of 5000 m. The electron microscopic investigation indicated that the majority of VLP were probably viruses.

Table 96-4 Abundances of bacteria and viruses in Nan-wan (Taiwan) samples enumerated with direct and film counts. Cells were collected with different filters; 0.2 μm Poretics (P), 0.2 μm and 0.02 μm Anodisc (A) and 0.015 μm Nuclepore (N)

Filter	Abundance at each depth (10^5 ml^{-1}) 0 m	5 m	10 m	20 m
Bacteria (direct count)				
0.2 μm P	5.22 ± 0.37	5.02 ± 0.19	4.33 ± 0.18	4.40 ± 0.41
0.2 μm A	5.01 ± 0.07	5.60 ± 0.17	5.23 ± 0.23	5.64 ± 0.27
0.02 μm A	5.30 ± 0.08	5.42 ± 0.38	5.82 ± 0.28	5.54 ± 0.16
0.015 μm N	4.76 ± 0.01	5.11 ± 0.26	5.75 ± 0.30	4.86 ± 0.26
Bacteria (film count)				
0.2 μm P	4.94 ± 0.03	4.94 ± 0.42	4.64 ± 0.18	4.44 ± 0.52
0.2 μm A	5.43 ± 0.30	5.42 ± 0.20	6.43 ± 0.31	5.66 ± 0.42
0.02 μm A	5.86 ± 0.13	5.04 ± 0.57	5.91 ± 1.10	4.99 ± 0.95
0.015 μm N	4.74	4.92	5.00	4.76
Viruses (film count)				
0.2 μm P	2.67 ± 0.43	2.38 ± 0.21	2.53 ± 0.27	2.59 ± 0.20
0.2 μm A	11.37 ± 4.85	12.76	11.23 ± 3.44	12.92 ± 2.93
0.02 μm A	4.91 ± 0.38	4.43 ± 0.93	4.55 ± 1.99	3.27 ± 0.79
0.015 μm N	21.63	18.56	17.61	17.57

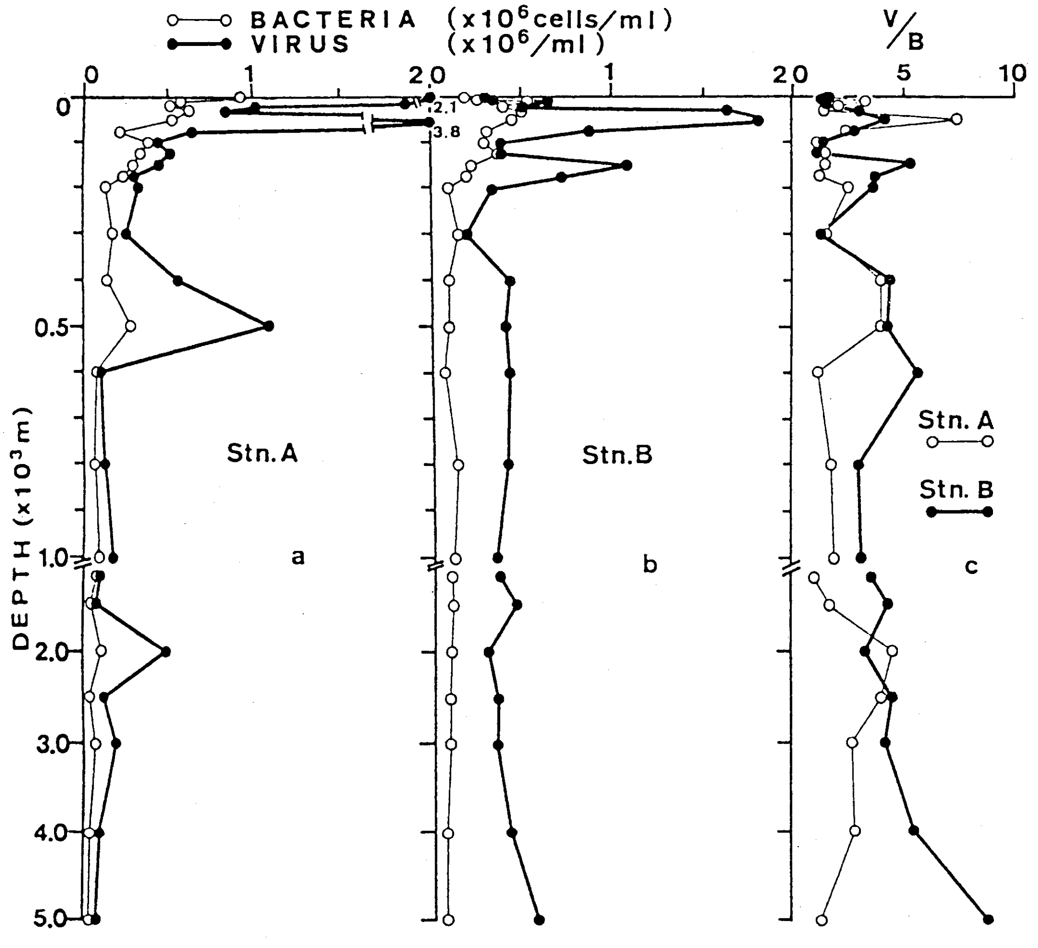

Fig. 96-29 Vertical distributions of bacteria and viruses at (a) Stn. A and (b) Stn. B, and (c) viruses/bacteria ratios at Stn. A and B.

5.4 Extraction and characterization of organic ligands from oceanic water columns by immobilized metal ion affinity chromatography

Midorikawa, and Tanoue (1996a)

Studies on the association of metals and organic compounds in natural waters have focused on metal speciation in situ. Our present understanding of copper speciation is that organic ligands exist in seawater and that more than 99% of Cu(II) in surface water is present in organic complexes.

Midorikawa and Tanoue (1996a) extracted organic ligands for Cu(II) from oceanic water columns using immobilized metal ion affinity chromatography (IMAC) (Fig. 96-30). Separation of organic ligands from bulk dissolved organic matter (DOM) enabled organic ligands to be chemically clarified. Measurements of complexing abilities and fluorescence and chemical analyses indicated that natural ligands were a mixture of, at least, two different types of organic ligands (Table 96-5). One type, prominent in surface water; was weakly fluorescent but rich in both primary amines and carbohydrates. The other type predominant in deep water, had converse characteristics, namely, low levels of both primary amines and carbohydrates, but relatively strong fluorescence. The measurement of organic ligand complexing ability from surface waters suggested the existence of a natural ligand ($\log K'_{CuL} \sim 9$) that has one or two primary amines as copper-binding sites.

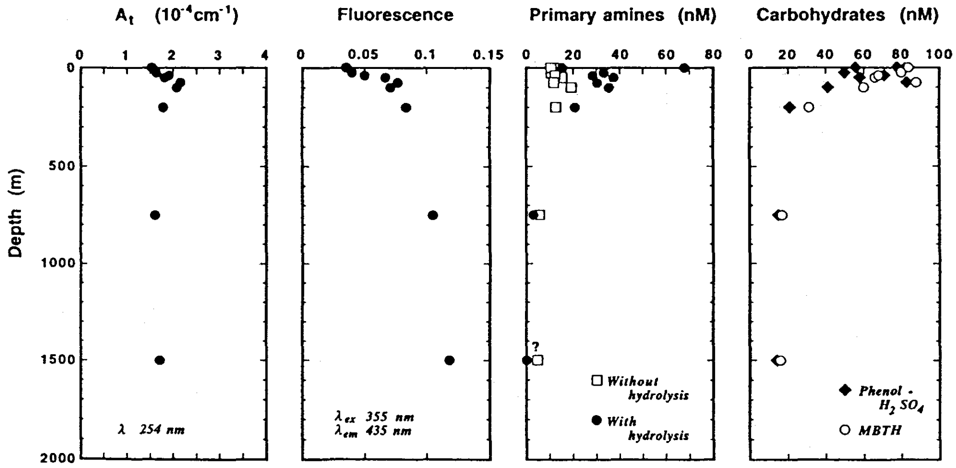

Fig. 96-30 Vertical profiles of organic-chemical characteristics of organic ligand fraction extracted by IMAC in equatorial Pacific. Water samples at Stations N-12 and M-2 were combined. Values are those in original seawater. Fluorescence is expressed in arbitrary units. Concentrations of primary amines with and without hydrolysis are given as nmol/l of glycine equivalents. The carbohydrate concentration was measured by phenol sulfuric acid and MBTH after hydrolysis and is given as nmol/l of glucose equivalents.
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Table 96-5 Results of copper titration and chemical analysis of the EDTA eluate of samples of surface seawater [a] after demetalization

Station	Column	Conditional stability constant [c]		Concentration (nM) [b]					A_t [b,f] (10^{-4} cm^{-1})	Recovery of UV absorbance [g] (%)
		log K'_{CuL_1}	log K'_{CuL_2}	Ligand [c]		Primary amine [d]	Carbohydrate [e]			
				C_{L_1}	C_{L_2}		P	M		
M-2	Upper	9.22	7.08	1.0	4.6	2.2	35	42	0.37	24
M-4	Upper	8.93	7.08	2.2	15	9.1	39	26 (?)	0.67	37
	Lower	8.92	7.44	0.5	3.1	3.8	17	16	0.16	25

[a] Seawater samples of 2.5 l (Station M-2) and 1.1 l (Station M-4) were each concentrated to titrated solutions of 10 ml.

[b] The respective values are expressed as those in the original seawater.

[c] The values represent values with respect to free copper under the conditions at $I = 0.7$ M (KNO_3), pH 8.15 (EPPS) and 25°C. The values for the ligand L_2 are preliminary because L_2 was detected but its precise quantification was hampered by the column blank.

[d] The values obtained without hydrolysis are given as nmol/l of glycine equivalents.

[e] The values were obtained by the phenol sulfuric acid method (P) and the MBTH method (M), and are given as nmol/l of glucose equivalents.

[f] The values for the total UV absorbance at 254 nm (A_t) of the EDTA-eluted fraction after demetalization were converted to those in the original seawater.

[g] The values were estimated as the ratio of the A_t value for the EDTA-eluted fraction after demetalization (i.e., ligand fraction with a molecular mass of more than 1000 Da) to that for the acidic eluate fraction without demetalization.

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5.5 Effects of ligand speciation on complexing abilities of strong ligands in natural waters

Midorikawa, and Tanoue (1996b)

The presence, in oceanic waters, of organic ligands (L) whose conditional stability constants (K'_{ML}) are strong enough to allow them to form complexes with copper has been reported, but no general consensus has been reached on the distribution of such strong ligands in the water column.

Midorikawa and Tanoue (1996b) found that these inconsistencies were derived from different analytical methods employed for their detection and different oceanographic conditions. In particular, the nature and quantity of detectable natural ligands are affected by the form the ligands are present in situ in different marine environments, that is, chemical speciation of natural ligands (ligand speciation), which depends strongly on the variations in concentrations of coexisting trace metals.

Using published data from observations in the laboratory and the field, they provided limits to the ranges of conditional stability constants of organic ligands for copper, zinc, and cadmium detectable by extensively used direct metal titration (Figs. 96-31 and 96-32). Their model indicates, for example, that organic ligands for copper with log $K'_{CuL(Cu')} > 12.4$ in surface water and with log $K'_{CuL(Cu')} > 9.9$ in deep water may not have been detected because stronger ligands formed complexes with ambient metals in situ at a station in the North Pacific.

The estimation suggests that there is a basin-scale difference in speciation of natural organic ligands and, moreover, that this difference influences metal speciation. They applied their ideas to more common oceanographic variations between the Pacific and Atlantic Oceans. There exist two remarkable differences in the respective vertical profiles of concentrations of total and inorganic copper in the two oceans: each level of copper in surface waters in the North Atlantic is not as greatly depleted as that in the North Pacific, whereas each copper level in the lower water column in the North Pacific is essentially about twice that in the North Atlantic (Fig. 96-33). Thus, these distributions have a major influence on ligand speciation in both oceans. In surface

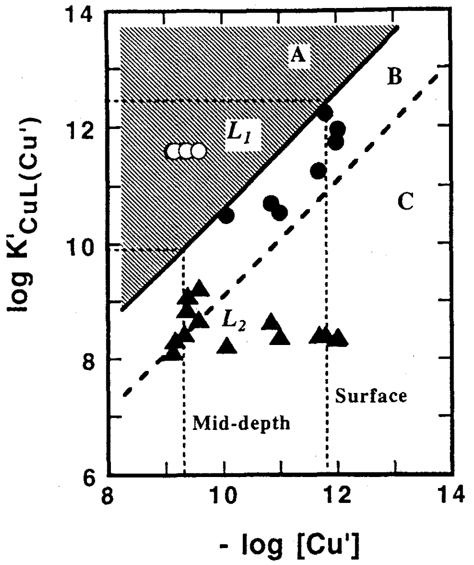

Fig. 96-31 [Cu'] and $K'_{CuL(Cu')}$ for ligands L_1 (circles) and L_2 (triangles) in original samples of seawater (20-1,400 m) from eastern North Pacific (VERTEX seasonal station: 33°N, 139°W): solid symbols: observed ligands; open symbols: hypothetical ligands (not observed: assuming lig $K'_{CuL1(Cu')} = 11.6$ for ligand L_1 at depths of 200-1,400 m). All data was from Coale and Bruland (1988). Solid line shows Eq. (5) for R = 80% and dashed line that for R = 10%; area A, $R > 80\%$; area B, $10\% < R < 80\%$; area C, $R < 10\%$. Dotted lines show average ligand speciation under conditions in surface water (pH 8.2 and 25 ℃) and at mid-depth (pH 7.5 and 5℃).

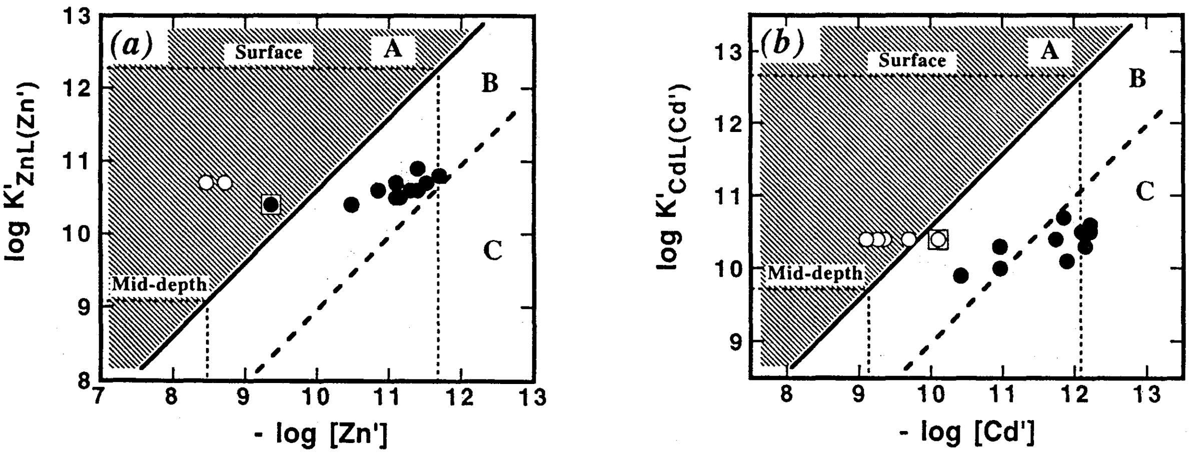

Fig. 96-32 Log $K'_{ML(M')}$ and -log [M'] for (a) zinc and (b) cadmium in original samples of seawater (22-600 m) from central North Pacific (VERTEX IV station: 28°N, 158°W) based on results of Bruland (1989, 1992): solid symbols: observed ligands; open symbols: hypothetical ligands (not observed; assuming log $K'_{ZnL(Zn')} = 10.7$ at depths of 500-600 m for zinc and log $K'_{CdL(Cd')} = 10.4$ at depths of 200-600 m for cadmium). Solid line shows Eq. (5) for R = 80% and dashed line that for R = 10%; area A, $R > 80\%$; area B, $10\% < R < 80\%$; area C, $R < 10\%$. Dotted lines show average ligand speciation under conditions in surface water and at mid-depth, calculated for each metal.

waters in the North Atlantic, in contrast with the North Pacific, the R for ligand L_1 and L_2 was more than 98% and that for ligand L_2 8-16%. (R is the percentage of complex between metal and ligand in total ligand concentration; R (%)/100 = $[ML]/C_L$) Thus, it could be predicted that the stronger ligand L_1 would be undetectable but the weaker ligand, corresponding to L_2, would be detectable in Atlantic surface water. It is postulated that comparisons of the occurrence and levels of organic ligands might not be possible among spatially and temporally different observations without reconciliation of the effects of ligand speciation, even if an identical method is used in all cases.

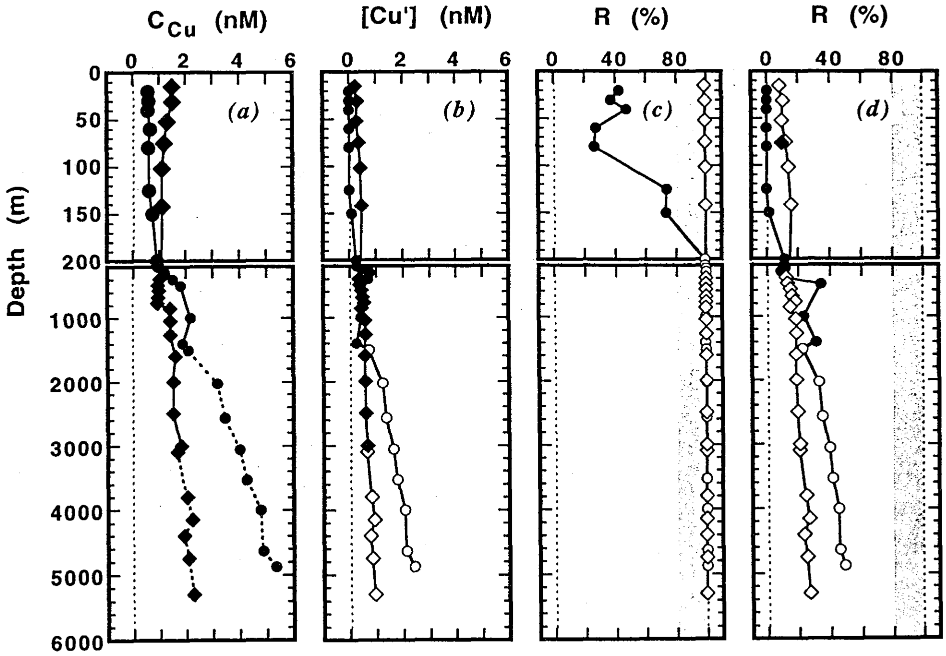

Fig. 96-33 Vertical profiles of copper concentrations and ligand speciation in North Pacific (circles) and the North Atlantic (diamonds): total copper (a), inorganic copper (b), R values for ligand L_1 (c), and L_2 (d). All solid symbols represent observed values and open symbols hypothetical, calculated values. Hatching shows the region (corresponding to area A in Fig. 96-31) in which the ligand cannot be detected by direct metal titration.

5.6 Determination of strong organic ligand dissolved in seawater: Thorium-complexing capacity of oceanic matter dissolved in oceans

Hirose (1996)

Oceanic organic matter, consisting of dissolved and particulate forms, is considered one of the important factors controlling the trace element composition of seawater and plays an essential role in in the marine environment carbon cycle. To understand the biogeochemical role of organic matter in the marine environment, it is important to specify and identify chemical constituents of oceanic organic matter. The composition of organic matter in seawater is extremely complicated. As a choice among many methodologies, the natural organic matter can be analyzed by functional group such as ligands related to metal complexation.

Hirose (1996) developed a way to measure strong ligands in oceanic DOM by Th complexation in acidic media and the adsorption of the Th complex onto XAD-2 resin. Th reacts quantitatively with the organic binding site of DOM in strong acid media (around 0.1 M H^+ solution), which is equilibrated within 24 hours. According to mass action analysis, Th forms a 1 : 1 complex with the binding site in DOM, whose conditional stability constant is $10^{6.7}$ M^{-1}. The conditional stability constant of the Th complex in DOM is in good agreement with that determined for oceanic particulate matter (PM) under similar experimental conditions.

This finding suggests that the chemical properties of the strong ligand in DOM are similar to these in biogenic PM. The Th complexing capacity in DOM, which corresponds to the total concentration of the strong organic ligand, can be determined (2-3 nM in surface waters) in a small volume (about 200 ml) (Fig. 96-34 and Table 96-6). The method has a detection limit of about 0.05 nM for thorium complexing capacity of DOM by using ^{230}Th.

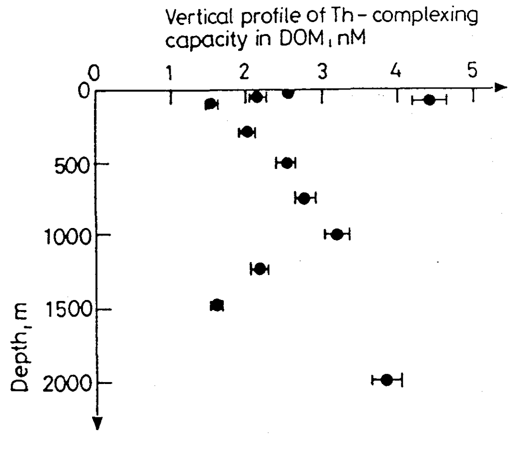

Fig. 96-34 Vertical distribution of Th complexing capacity of DOM in Japan Sea. (Sampling site: 38°11′N, 132°37′E)

Table 96-6 The vertical distribution of Th complexing capacity in DOM. (Sampling location: 38°11′N, 132°37′E; depth: 2354 m; sampling date: Oct. 1994)

Depth, m	T, °C	S, ‰	ThCC, nM
0	19.6	33.42	2.56 ± 0.10
50	17.6	33.67	2.13 ± 0.10
75	9.95	34.23	4.41 ± 0.13
100	6.02	34.13	1.53 ± 0.06
300	0.76	34.07	2.00 ± 0.08
500	0.40	34.07	2.51 ± 0.13
750	0.27	34.07	2.77 ± 0.12
1000	0.21	34.07	3.18 ± 0.16
1250	0.18	34.07	2.16 ± 0.11
1500	0.16	34.07	1.59 ± 0.06
2000	0.17	34.07	3.86 ± 0.14

Uncertainties quoted are at a level of one estimated standard deviation for counting only.

INTERNATIONAL ACTIVITIES

II. Participation in At-Sea Intercomparison of Underway pCO_2 Systems During *Meteor* Cruise 36-1

H. Y. Inoue and M. Ishii participated in the At-Sea Intercomparison of Underway pCO_2 Systems during German research vessel *Meteor* Cruise 36-1, June 6-19, 1996, organized by the marine CO_2 project at the Institute for Marine Research, Department of Marine Chemistry, Kiel, Germany. The purpose was to provide insights into the performance and comparability of seagoing CO_2 systems under typical identical working conditions to establish a database for use in understanding the basin and global scale distributions of pCO_2 and its influence on the oceanic uptake of anthropogenic CO_2.

Fifteen scientists from nine research institutions in six countries joined the international exercise, conducted on board R/V *Meteor* between Bermuda and Gran Canaria in the North Atlantic. H.Y. Inoue and M. Ishii contributed to the intercomparison exercise using the underway MRI pCO_2 system.

III. Contribution to WOCE Hydrographic Program — Pacific Data QA Activity

M. Aoyama was engaged in data quality evaluation (DQE) of Pacific WOCE data as an evaluator with the region study to assess data quality and make suggestions for improving it. He worked for the WHP Office in the Physical Oceanography Department of the Woods Hole Oceanographic Institution as a guest investigator from March 1, 1996 to May 31, 1996.

Aoyama applied original methods and compared data at crossing one-time survey lines in the North Pacific to assess the degree to which data is selfconsistent and examined individual datasets for data quality.

Details of his contribution to the Pacific Data QA Activity are given in the WHP Office Status Report, No. 19.

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気象研究所

1946（昭和21）年 設立

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気象研究所技術報告の編集は，編集委員が行う。編集委員会は原稿の掲載の可否を判定する。

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