Temporal variations of heavy metals in coral *Porites lutea* from Guangdong Province, China: Influences from industrial pollution, climate and economic factors *

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Abstract The eight heavy metals Cr, Mn, Co, Ni, Cu, Zn, Cd, and Pb have been determined in samples of coral *Porites lutea* collected from Dafangji Island waters (21°21'N, 111°11'E), Dianbai County, Guangdong Province, China, by the ICP-MS method. The samples represent the growth of coral in the period of 1982-2001. The results showed that the waters were polluted by the heavy metals Cu, Ni, Zn, and Pb in certain years, but not by other metals. The contamination may have come from industrial sources, including electroplating, metallurgy, mining, and aquatic industries in the coastal areas.

The correlation coefficients among the metals and climatic and economic factors indicate that the metals Ni, Zn, and Cd behave similarly. Copper and Mn are positively correlated, and cobalt is negatively correlated with Cr, Ni, Zn, and Cd. Lead is not correlated with any other metals but is correlated with sea surface water temperature, air temperature, GDP and industrial-agricultural production in Dianbai County. Lead in corals is related to the enhanced pollution level of ocean waters as a result of increased industrial activities.

Key words coral *Porites lutea*; heavy metal; oceanic pollution; climate and economic factors

1 Introduction

Corals are an important proxy for reconstructing paleo-environment and revealing global changes in the past. Variations of heavy metals in the growth bands of corals can provide important information about the oceanic environment.

Bastidas and Garcia (1999) analyzed ten metals (Al, Ca, Cd, Cu, Fe, Cr, Hg, Pb, V and Zn) in skeleton sections of coral *Porites astreoides* from Venezuela and found that the skeletal Al, Fe, Cr and Ca contents varied significantly in relation to distances of sampling localities, to river inputs, and to growth rates of corals from different locations. Reichel-Brushett and Harrison (1999) studied the effects of Cu, Zn, and Cd on fertilization of gametes from reef coral scleractinian and showed that copper can be toxic to the fertilization process of gametes from brain corals. Miao et al. (2001) measured As, Cd, Cr, Cu, Pb, Hg, Se and Zn in corals, crab, fish, lobster and sediments collected from French Frigate Shoals and found that these metals in the organisms and sediments did not vary so much. It does not seem that human activity was a factor leading to the accumulation of metals, with the possible exception of lead. David (2003) studied the historical input of mine tailings into the western coastal ocean in terms of trace metal variations in the growth bands of coral *Porites* from the Marinduque Island, Philippines, which reflected the contamination of the surrounding area with higher Cu, Fe, Mn, and Zn concentrations derived from the mine tailings. Increased metal concentrations in corals are due to their proximity to a tailing source and/or due to increased erosion/sedimentation in the coastal area with high rainfall. Liu Chunying et al. (2001) and Yu Kefu et al. (2002) determined the heavy metals Cu, Pb, Cr in coral *Platygyra* from the Daya Bay, China, and found that the ecological environment there was not influ-

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enced by heavy metals during 1976 – 1998.

We have collected samples of coral *Porites lutea* from the South China Sea region and carried out geochemical analysis to obtain paleoclimatic information since 1997. Peng Zicheng et al. (1998, 2003a) performed TIMS-U series disequilibrium dating of corals and discussed the variation of the sea level in the South China Sea in the last 7000 years. Liu Weiguo et al. (1999) measured $\delta^{11}B$ and boron content in coral *Porites lutea* from the South China Sea and their values are related to the sea surface pH values and CO_2 concentrations. He Xuexian et al. (2002) investigated the relationship between $\delta^{18}O$ and sea surface temperature (SST) using coral *Porites lutea* from the Hainan Island waters, and found the periodic peaks of the SST series at 2 – 2.5 years (QBO band) and 3 – 7 years (ENSO band). Peng Zicheng et al. (2003b) used the records of $\delta^{18}O$ in corals as an indicator of winter monsoon intensity in the South China Sea, and obtained a 54-year-long calculated winter monsoon intensity index (WMII) series. The general tendency of the WMII series shows a decrease from 1944 to 1998, which may be related to global warming.

The purpose of this paper is to describe the variation trend of heavy metals in coral *Porites lutea* from the Dafangji (DFJ) Island waters, Guangdong Province, which is situated to the north of the South China Sea, and to investigate the influence of industrial pollution and their relationships to climate and economic factors.

2 Sampling site

In August 2002, a massive sample of coral *Porites lutea* was collected underwater at a depth of 9 m in the Dafangji (DFJ) Island waters (21°21′N, 111°11′E), Dianbai County, Guangdong Province, China. The top part of the coral was still alive when the sample was collected.

The DFJ Island area has a South Asian tropical monsoon climate, with a high temperature and high precipitation in summer seasons, and with a lower temperature and less precipitation in winter seasons. Climate records from the Dianbai Observatory Station showed that the annual mean air temperature is 23.0℃ (1961 – 1990). The lowest mean monthly temperature is recorded in January (15.7℃) and the highest in July (28.6℃). The annual mean rainfall is 1520 mm (1961 – 1990) and the rainfall is concentrated in the months from April to September. The winter monsoon prevails from November to February with the east wind due to the influence of coastal terrain. The summer monsoon prevails from March to August with the south wind. The monthly mean wind speed is 2.0 – 3.2 m/s. The sea surface temperature (SST) data filed by the Naozhou Observatory Station (20° 56′N, 110° 35′E) showed that the SST increased at a rate of +0.58℃/10 a during 1982 – 2001 ($n = 20$, $r = 0.63$), which may be related to global warming.

3 Methods

The coral sample was cut into 5-mm thick sections by means of a steel saw blade. Each section was washed with pure water and immersed in Mill-Q water (18.2 MΩ/cm) over night, then it was cleaned three times in an ultrasonic bath and washed again with Mill-Q water. Finally, the section was dried in an oven at 50 – 60℃ for taking X-ray image. The X-ray photograph shows the structure of corals in density-band couples (DBC) at a resolution 0.5 – 1 year/sub-sample (Fig. 1). A total of 23 samples, corresponding to the

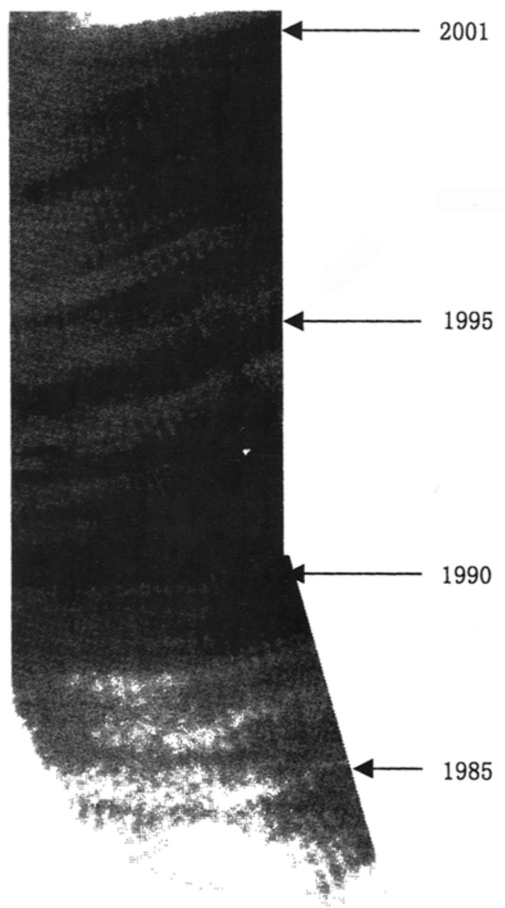

Fig. 1. X-ray radiograph of coral *Porites lutea* in 1982 – 2001 from the Dafangji Island waters, Dianbai County, Guangdong Province, China. High- and low-density band couplets at annual resolution are well developed.

years 1982 – 2001, were prepared.

The coral sub-samples were dissolved in hydrochloric acid and then analyzed for heavy metals such as Pb, Cd, Cu, Cr, Co, Mn, Ni, Zn using the VG-PQ3 ICP-MS technique. The measurement procedures are described as follows. First, one solution composed of elements of the same concentration (1.0 μg/L) but different mass numbers (Be, Co, In, Bi, U) was used to adjust the working parameters in order to get the maximum isotopic count rate in the mass range of 9 – 238 AMU. The isotopes ^{208}Pb, ^{111}Cd, ^{65}Cu, ^{52}Cr, ^{59}Co, ^{55}Mn, ^{60}Ni and ^{66}Zn were chosen to be analyzed because these nuclides have high abundances, little interference and low detection limits. Blank and standard solutions, containing 10 μg/L of each of the elements Pb, Cd, Cu, Cr, Co, Mn, Ni and Zn, were used to establish standard curves. The ^{115}In solution (40 μg/L) was used as an internal standard and was mixed thoroughly with the solution of sub-sample in order to eliminate the effect of equipment drift so as to enhance the precision of the measurement. The precision of the methods was determined by replicate analyses of a standard solution (Table 1). The relative standard deviations are between 1.4% and 4.9%.

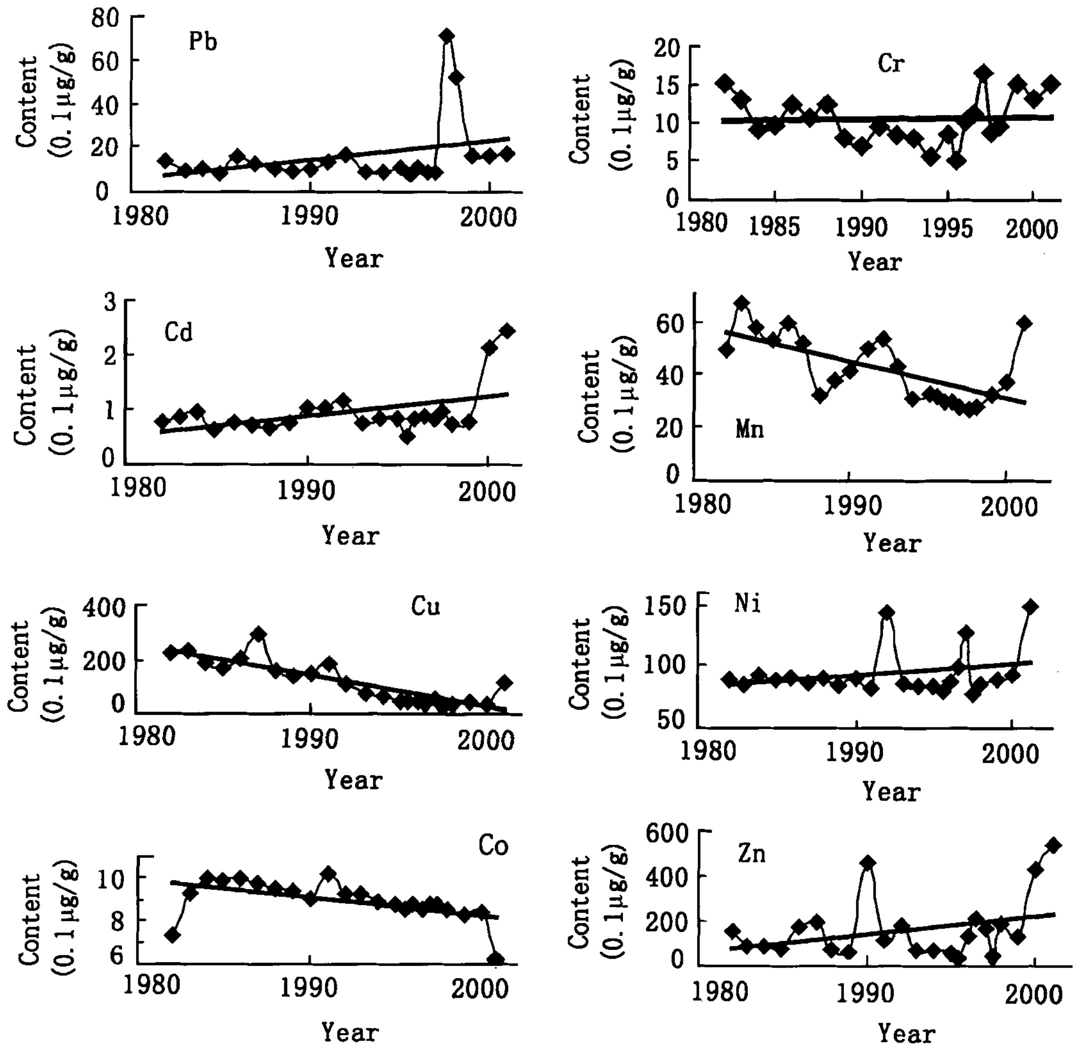

Fig. 2. The yearly variations of heavy metals in the Dafangji coral, Dianbai, Guangdong, during 1982 – 2001. The slope of correlation lines is 0.094 μg/a for Pb, 0.004 μg/a for Cd, –1.2 μg/a for Cu, –0.008 μg/a for Co, 0.005 μg/a for Cr, –0.13 μg/a for Mn, 0.10 μg/a for Ni, and 0.86 μg/a for Zn, respectively.

4 Results

The abundances of the eight heavy metals in coral samples are listed in Table 2. The Pb contents of corals vary from 0.75 – 7.24 μg/g with an average of 1.62 μg/g (n = 23). The values are approximate to the range of 0.5 – 8.75 μg/g with an average of 1.57 μg/

g ($n = 55$) in corals from the Daya Bay (Liu Chunying et al., 2001). The Pb variation at yearly intervals shows a peak on May 1997 and 1998 (Fig. 2). The peak values are 2.3 - 3.5 times higher than the average value, which implies that the Pb pollution had happened in the DFJ Island waters during these years. The slope of the regressive line indicates the Pb contents of the corals increased at a rate of 0.09 μg/g per year during the period from 1981 to 2001.

Table 1. Precision of the method determined by replicate analysis of a standard solution using the VG-PQ3 ICP-MS equipment

	Pb	Cd	Cu	Co	Cr	Mn	Ni	Zn
Standard concentration (μg/g)	40.00	40.00	40.00	40.00	40.00	40.00	40.00	40.00
Mean of the measured results ($n = 13$, μg/g)	39.95	40.01	40.05	39.90	39.83	40.06	39.96	40.39
Standard deviation (μg/g)	0.54	0.68	1.39	1.2	1.66	1.41	1.41	1.98
Relative standard deviation (%)	1.4	1.7	3.4	3.0	4.9	3.5	3.5	4.9

Table 2. The abundances of the eight heavy metals in the Dafangji coral, Dianbai County, Guangdong Province, China, determined by the ICP-MS method (in 0.1 μg/g)

Sample No.	Year	Sample weight (g)	Pb	Cd	Cu	Co	Cr	Mn	Ni	Zn
DFJ-1-1	2001	0.07454	18.08	2.49	121.7	6.29	15.61	61.54	151.1	551.7
DFJ-1-2	2000	0.08606	17.39	2.19	37.33	8.45	13.64	37.91	95.63	447.3
DFJ-1-3	1999	0.08273	17.00	0.84	42.34	8.33	15.54	33.24	90.99	139.9
DFJ-1-4	1998	0.09331	53.42	0.76	34.96	8.63	9.91	29.2	86.94	197.6
DFJ-1-5 (1)	1997.5	0.07060	72.41	0.99	28.24	8.89	9.10	27.61	79.46	53.4
DFJ-1-5 (2)	1997	0.07608	9.01	0.86	52.28	8.82	16.95	29.22	129.3	183.2
DFJ-1-6 (1)	1996.5	0.08028	9.15	0.91	36.50	8.65	11.58	30.24	99.93	220.7
DFJ-1-6 (2)	1996	0.07270	10.83	0.85	51.10	8.81	10.5	30.65	88.55	149.3
DFJ-1-7 (1)	1995.5	0.08278	7.46	0.54	45.21	8.64	5.32	32.31	80.00	42.04
DFJ-1-7 (2)	1995	0.08610	11.07	0.84	48.34	8.83	8.77	33.77	84.38	70.01
DFJ-1-8	1994	0.07658	8.84	0.86	64.31	8.95	5.79	31.32	85.04	75.02
DFJ-1-9	1993	0.07910	9.21	0.76	74.84	9.34	8.21	43.99	86.09	74.46
DFJ-1-10	1992	0.07686	17.26	1.2	114.4	9.34	8.59	55.00	145.8	188.4
DFJ-1-11	1991	0.08266	13.81	1.1	189.4	10.26	9.71	50.66	82.90	127.5
DFJ-1-12	1990	0.08228	10.63	1.1	149.4	9.12	7.08	41.93	91.03	461.8
DFJ-1-13	1989	0.08638	8.64	0.77	139.5	9.42	8.14	38.17	84.16	67.52
DFJ-1-14	1988	0.07677	9.62	0.68	157.4	9.60	12.59	32.37	90.40	72.29
DFJ-1-15	1987	0.07997	12.39	0.72	296.5	9.84	10.81	52.61	86.88	204.9
DFJ-1-16	1986	0.07980	15.29	0.75	201.5	10.07	12.54	60.46	91.35	178.4
DFJ-1-17	1985	0.05680	7.72	0.65	165.6	9.92	9.69	53.96	89.00	75.18
DFJ-1-18	1984	0.09174	10.29	0.97	185.0	10.02	9.15	58.45	92.60	85.16
DFJ-1-19	1983	0.07969	9.34	0.88	233.7	9.33	13.10	68.52	84.77	85.61
DFJ-1-20	1982	0.05430	13.59	0.78	221.3	7.33	15.24	49.82	89.18	157.1
Range/Mean ($n = 23$)			7.5-72.4/16.2	0.54-2.49/0.97	28.2-297/117	6.3-10.3/9.0	5.3-17.0/10.8	27.6-68.5/42.7	79.5-151/95.0	42.0-551/169
Range/Mean from Liu Chunying et al. (2001)			5-85.7/15.7 ($n = 55$)	0.3-5.2/1.1 ($n = 43$)	50-66.2/25.7 ($n = 56$)					
Range/Mean ($n = 115$) from Guo Lifen et al. (1997)			38-1790/233	0.5-120/8.2	12.5-880/202	0.1-190/31.4	1.40-237/58.6	12.4-2260/164	10-640/52	11.8-960/172

The Cd contents of the corals vary over a range of 0.054 - 0.25 μg/g with an average of 0.097 μg/g ($n = 23$), also similar to a range of 0.03 - 0.52 μg/g with an average of 0.11 μg/g ($n = 43$) in corals from the Daya Bay. The yearly variation of Cd shows corals were enriched in Cd in the years of 2000 and 2001. The Cd contents of corals in 2000 were 2.3 times the average value, and those in 2001 were 2.6 times the average value. The regressive line indicates a slightly increasing tendency of Cd content at an annual rate of

0.004 μg/g during 1982 – 2001 (Fig. 2). The results revealed that the Cd pollution to a certain extent happened in recent years (Fig. 2).

The Cu contents of the corals vary from 2.82 to 29.7 μg/g with an average of 11.7 μg/g, which are higher than those of corals from the Daya Bay over the range of 5.0 – 6.62 μg/g with an average of 2.57 μg/g ($n = 56$). The highest Cu content occurred in 1987, 2.5 times the average value. The copper contents generally decreased at an annual rate of 1.2 μg/g during 1981 – 2001 (Fig. 2).

The contents of other metals such as Co, Cr, Mn, Ni and Zn have a range and an average corresponding to 0.63 – 1.03 μg/g and 0.9 μg/g for Co, 0.53 – 1.7 μg/g and 1.08 μg/g for Cr, 2.76 – 6.85 μg/g and 4.27 μg/g for Mn, 7.95 – 15.1 μg/g and 9.54 μg/g for Ni, and 4.2 – 55.1 μg/g and 16.9 μg/g for Zn, respectively. The Cr contents of the DFJ coral are consistent with those of the Ximao coral, while Ni and Zn in the DFJ coral are obviously higher than those in the Ximao coral. Most of the heavy metals in the DFJ coral fall within the range of the metal contents of corals from Nansha reefs (Table 2, Guo Lifen et al., 1997). Guo Lifen et al. (1997) pointed out that the higher contents of heavy metals such as Pb, Cd, Cu, Cr, Mn, etc in the Nansha reef-corals are due to the influences from continental dross, the exploitation of oil fields, navigation garbage and oil leakage, which are harmful to the ecological environment in the Nansha waters of the South China Sea.

Yearly variations of the Co, Mn, Cr, Ni and Zn contents in corals are shown in Fig. 2. Cobalt level in 1991 and Mn level in 1983, 1986, 1992 and 2001 are shown as small peaks; their values show slight enhancement but are 1.6 times less than the respective averages. It seems there is no serious pollution with respect to Co and Mn. The Co and Mn contents decreased at the rates of 8 μg/g and 133 μg/g per year, respectively during 1981 – 2001. Chromium shows a moderate enrichment in 1982, 1997, and 1999 – 2001, but the values are only 1.4 times less than the average Cr content. The Ni contents have an average of 9.5 μg/g, which exceed those of the Nansha and Ximao corals. The average of the Zn contents is 16.9 μg/g, which is close to 17.2 μg/g for the Nansha coral. The Ni peaks appeared in 1992, 1997 and 2001 with the values being 14.6, 12.9 and 15.1 μg/g respectively, 1.4 – 1.6 times the average value. Otherwise the Zn peaks appeared in 1990 and 2000 – 2001 and the corresponding values are 2.8 – 3.3 times the average value. It may be noted that both Zn and Cd enrichments appeared in the years of 2000 and 2001, reflecting their geochemical coherence.

The results showed that waters off the Dafangji Island area were contaminated by the heavy metals Zn and Ni to a certain extent. That seems to be caused by electroplating, metallurgy, mining and aquatic industries in the coastal region. It is worth to be pointed out that the yearly variations of the 8 heavy metals show different patterns (trends and peak-years), indicating that the heavy metals may come from different sources. It is necessary to make a comprehensive plan for the prevention of heavy metal pollution.

5 Heavy metals in corals in relation to climate and economic factors

The temporal variation of heavy metals in the corals is related to seawater pollution. Coral is a living organism, and absorbs nutrients and metabolizes during its growth. During the formation of coral skeletons of calcium carbonate other chemical elements including heavy metals are also incorporated. The distribution of chemical elements between corals (aragonite) and seawater not only obeys the geochemical principles, but also depends on biochemical and physiological controls. Therefore, heavy metals in the corals can be related to climatic factors such as sea surface temperature (SST), air temperature (AT) and sunshine irradiation (IR). Moreover, the heavy metal contents of corals from the Dafangji Island can also be related to the gross domestic product (GDP) and industrial-agricultural production (IAP) of Dianbai County, which represent the economic conditions related to the pollution level to a certain extent.

The correlation coefficients among the heavy metals, climatic and economic factors are calculated using software SPSS10.0 (Table 3). The confidence levels of positive or negative correlation coefficients are at 95%, and those of strongly positive or strongly negative correlation coefficients are at 99%.

Lead in the Dafangji corals is positively correlated with SST, IAP and GDP and is strongly correlated with AT, which indicates that at higher air temperature and higher SST, more Pb will be dissolved in the seawater, and consequently more Pb will be absorbed by corals. Furthermore, when IAP and GDP are on the rise, more industrial wastes containing Pb are generated, which may cause an increase in dissolved lead in the seawater and subsequently in coral skeletons.

Cadmium shows a positive correlation with Ni and Zn. These three metals may be derived from a common source (s) because of similar temporal variation trends (Fig. 2). Cadmium has a strongly negative correlation with Co, which indicates the two heavy metals may be derived from different sources. The highest content of

Cd appeared in 2001, but the lowest value of Co appeared in the same year.

Table 3. Correlation coefficients among the heavy metals, climatic and economic factors

Element	Positive correlation	Strongly positive correlation	Negative correlation	Strongly negative correlation
Pb	SST (0.548)	AT (0.626)		
	IAP (0.548)			
	GDP (0.572)			
Cd		Ni (0.656)		Co (-0.586)
		Zn (0.823)		
Cu		IR (0.658)		IAP (-0.873)
		Mn (0.733)		GDP (-0.845)
Co			IAP (-0.531)	
			GDP (-0.533)	
			Cd (-0.586)	
			Cr (-0.510)	
			Ni (-0.481)	
			Zn (-0.550)	
Cr			Co (-0.510)	
Mn	IR (0.478)	Cu (0.733)		IAP (-0.741)
				GDP (-0.710)
Ni	Zn (0.534)	Cd (0.656)	GR (-0.540)	
			Co (-0.481)	
Zn	Ni (0.543)	Cd (0.823)	Co (-0.550)	

Note: The sea surface temperature (SST) records in 1982 - 2001 from the Naozhou Observatory Station (20° 56′N, 110° 35′E); the air temperature (AT) and sunshine irradiation (IR) records in 1982 - 2001 from the Dianbai Observatory Station; the gross domestic product (GDP) and industrial-agricultural production (IAP) in 1982 - 2001 from the Dianbai County Government. The Pearson correlative coefficients are calculated using software SPSS10.0, and are listed in the parentheses.

Copper is strongly positively correlated with Mn and IR. Copper and Mn may be derived from a common source. Also an increase in sunshine irradiation may help corals absorb more Cu and Mn. Interestingly, copper is negatively correlated with IAP and GDP, indicating that Cu and Mn uses decreased in correspondence to a trend of increasing GDP and IAP of Dianbai County during 1982 - 2001. It can be inferred that the exploitation of Cu and Mn deposits and application of the two metals to industrial production showed an atrophy during that period of time.

The Co contents vary within a narrow range of 0.83 - 1.0 μg/g for all the years except 1982 (0.73 μg/g) and 2001 (0.62 μg/g). Cobalt is negatively correlated with IAP, GDP, Cd, Cr, Ni and Zn. So the behavior of Co related to the industrial factors (IAP, GDP) is similar to that of Cu and may be derived from different sources of Cd, Cr, Ni, and Zn.

Chromium is negatively correlated with cobalt and has no significant correlation with other elements or industrial and environmental factors.

6 Conclusions

The eight heavy metals Pb, Cd, Cu, Co, Mn, Ni, and Zn in coral *Porites lutea* from the Dafangji Island waters, Dianbai County, Guangdong, China, were determined using the VG-PQ3 ICP-MS technique. The samples represent the growth period of corals during the years from 1982 to 2001. Temporal variations of heavy metals show that if the contents of a heavy metal are 2 times less than its average value, presumably the coral is believed not to be polluted or only is subjected to slight pollution. If the contents of a heavy metal in corals are more than 2 times the average value, the corals are likely to have been polluted. If the metal contents are more than 3 times the average value, the corals have obviously suffered serious pollution. At present, there is no national standard to judge whether a piece of coral is contaminated by heavy metals. Also, if the average value of a heavy metal in the coral is much higher than that of corals from other areas in China, it may be suggested that the coral has been polluted to a certain extent.

The average Ni and Zn contents in the Dafangji corals are very high as compared with those of corals from other areas in China. Moreover, the contents of Zn in 1990 and 2001, Pb in 1997 and 1998, and Cu in 1987 are 2 - 5 times the respective averages. The Dafangji corals were conspicuously polluted by the heavy metals Zn, Pb, Cu in certain years. The pollutants may be derived from electroplating, metallurgy, mining, and aquatic industries along the coastal areas.

Correlation coefficients among the heavy metals, and the climatic and economic factors were calculated. It is shown that Cd, Ni, and Zn have similar behaviors. Copper behaves like Mn, and cobalt is negatively correlated with Cr, Ni and Zn. Lead is not correlated with other metals. The heavy-metal contents of the Dafangji corals appear to be related to the climatic and economic factors.

The temporal variations of heavy metals in corals are not only caused by the pollution alone. Coral is a living organism, and the abundances of the elements in coral skeletons are influenced by the biochemical and physiological characteristics of corals. Therefore, in making a plan to prevent heavy-metal pollution along the coastal areas, a holistic approach is needed.

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