### GEOTRACES-CHINA GPpr15-summer Process Study Cruise Report

R/V Tan Kah Kee (KK2003)

July 3, 2020 - August 20, 2020

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August 20, 2020

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#### 1. Executive Summary

The GEOTRACES GPpr15/KK2003 Cruise was conducted aboard R/V TAN KAN KEE" ("嘉庚" 号) from July 3<sup>rd</sup> to August 20<sup>th</sup>, 2020, in the western North Pacific Ocean. The cruise was led by the State Key Laboratory of Marine Environmental Science of Xiamen University (MEL), and was joined by scientists from Ocean University of China (OUC) and Shanghai Jiaotong University (SJTU). It is supported by a "Ocean Desert" Major Project led by Dr. Minhan Dai sponsored by the National Natural Science Foundation of China (No. 41890800).



Figure 1. GPpr15/KK2003 Cruise study map

The oligotrophic ocean occupies about 30% of the ocean surface and has been conventionally regarded as ocean deserts. It is characterized by nutrient depletion in the surface waters and extremely low net biological production and hence, per unit area, contributes little to carbon export from surface to deep waters. Emerging evidence, most notably based on ocean time-series studies such as those at the Hawaiian Ocean Time-series station, has shown a wider than previously assumed dynamic range of nutrient inputs and biological responses in this oceanic system. This project selects the North Pacific Subtropical Gyre (NPSG), one of the world's largest oligotrophic regimes, as the study site to examine carbon fixation and export, or the biological pump in general, regulated by differently sourced nutrients including macronutrients (i.e., N, P, Si) and micronutrients (e.g., Fe).

In 2019, the first CHINA- GEOTRACES cruise (GP09/KK1903), a pilot cruise of Carbon-FE was conducted from April 25 to June 13 in the western North Pacific (wNP). We have obtained the spatial distributions of temperature, salinity, dissolved oxygen, nutrients, and dissolved iron (DFe)

in the wNP. Underway measurements of DFe, ranging between 0.17 and 0.54 nmol  $L^{-1}$ , showed a decreasing trend from northwest to

southeast of the wNP. Turbulent microstructure observations showed that the turbulent kinetic energy dissipation rate varied within a range of  $1.0 \times 10^{-10}$  to  $1.0 \times 10^{-5}$  W kg<sup>-1</sup>, which will be used to calculate the nutrient diffusive fluxes. Nutrient addition experiments suggested that nitrogen (N) was the major limiting factor of phytoplankton growth in the wNP. Preliminary results from sediment traps showed that sinking particles in the nutrient depleted layer (NDL) had lighter N isotopic compositions, suggesting that N<sup>2</sup> fixation is the major source of N nutrient in the NDL. In contrast, the subsurface supply should be the primary nutrient source to the nutrient replete layer, support the export production therein.

Therefore, the GPpr15 process was further designed to explore the biogeochemistry of trace elements and isotopes (TEIs) in the wNP, covering the study area between 10-33°N and 118.5-155°E with a set of 21 stations, including 5 Mega Stations, 14 Clean Stations, and 3 Normal Stations, with trace metal clean surface towed fish sampling in 162 stations (for concentrations and isotopic compositions of Fe, Ni, Nd, etc) (Fig. 1). The main purposes of the GPpr15/KK2003 Cruise are as follows:

- (1) To determines the distribution of macro- and micro-nutrients, fingerprint their sources, and estimate their fluxes into the NPSG
- To constrain the spatial-temporal variability of biological N<sup>2</sup> fixation and its limiting factors in the NDL,
- (3) To quantify the carbon fixation and associated planktonic community structure,
- (4) To constrain the export production from both the NDL and NRL,
- (5) to simulate the biological pump and carbon sinks in the NPSG.

During the 51 days cruise, seawater samples were collected with a conventional CTD and a trace metal clean CTD, respectively, and near-surface underway seawater samples were collected from a towed fish sampling system for TEIs, nutrients, pigments, and on-deck incubations. The cruise also conducted trace metal clean in situ pump for trace metal concentrations on particles in 9 stations (89 samples in depths ranging from 25-3500 m) by McLane In-Situ Pumps with a SBE37IM CTD attached to each pump. Seawater, particles, and aerosol samples were collected in this cruise for the determination of TEIs. GEOTRACES key trace elements and isotopes in collected samples, including some other elements and isotopes REEs, <sup>234</sup>Th, <sup>210</sup>Pb-<sup>210</sup>Po, and silicon and barium isotopes, will be analyzed after the cruise in shore-based laboratories.

Regular CTD casts in 17 stations with the others physical (salinity, currents, turbulence, etc), chemical (DO, DIC, POC, macro-nutrients, etc), and biological (chlorophyll *a*, pigments, flow cytometry and molecular samples etc.) parameters. Turbulence dissipation rate, hydrological parameters (T, S, Turbidity, etc), and meteorological parameters were measured at stations and/or underway. Nutrient profiles, including nanomolar level phosphate and nitrate in shallow water column, were analyzed aboard for each station. A series of on-deck incubation experiments were also conducted with emphases on the interactions between ecosystem functions and macro- and micro-nutrient supply. These incubation experiments were conducted for phytoplankton primary

production, nitrogen fixation, bacterial production and respiration, zooplankton grazing rates, etc. In addition, Lagrange observations were conducted at 3 Mega stations by surface floating drifts and 3 sediment traps in both the NDL and NR.

This project can substantially improve our understandings to fundamental biogeochemistry in these climatically and ecologically important oligotrophic ocean systems.

We sincerely acknowledge Captain Long Yin, the crew, and the marine technical support team of the R/V Tan Kah Kee for their invaluable and successful support of all shipboard operations. Sincere thanks are also given to the Office for Research Vessel Operation Center of Xiamen University for its flawless logistic support to the cruise. We specially thank Liping Zhou (Peking University) and the whole international GEOTRACES community for their continual scientific advices and technical instructions to make the cruise possible.

### 2. Principle Programs

Paramatars	In-charge Person	PI name	Submission
	In-charge rerson		Date
Trace metals			
FIA Fe	Yongming Huang	Yihua Cai	2021-12
Fe (ICP-MS)	Liping Ye	Yihua Cai	2021-12
Cu, Zn, Cd, Ni, Pb, Co	Junbo Yang	Yihua Cai	2021-12
Dissolved and particle <sup>210</sup> Po	Xinru Zhang	Yihua Cai	2021-12
Dissolved and particle <sup>210</sup> Pb	Xinru Zhang	Yihua Cai	2022-6
Nd	Wenkai Guan	Jin Zhang	2022-6
Fe isotope	Ziyuan Jiang	Ruifeng Zhang	2023-6
Particle trace metal	Kan Zhang	Minhan Dai	2021-6
Total Hg/CH <sub>4</sub> -Hg	Wanyang He	Jing Zhang	2021-6
Fe concentration	Zhan Shen	Ruifeng Zhang	2021-8
REEs	Wenkai Guan	Jing Zhang	2022-6
Al	Wanyang He	Jing Zhang	2021-12
DIC-13C, DIC-14C	Ying Gao	Minhan Dai	2021-3
BSi/PBa Trap&pump	Xinting Rao	Zhimian Cao	2021-8
DSi/DBa	Xinting Rao	Zhimian Cao	2021-8
BSi/PBa	Xinting Rao	Zhimian Cao	2021-8
Total 234Th	Yifan Ma	Minhan Dai	2021-8
Particle 234Th, POC	Yifan Ma	Minhan Dai	2021-8
Trap POC/PN	Yifan Ma	Minhan Dai	2021-8
Hydrography			
CTD		Zhiyu Liu	Submitted
ADCP		Zhiyu Liu	Submitted
MVP		Zhiyu Liu	Submitted
Underway T and S, AWS		Zhiyu Liu	Submitted
Underway Chl a	Zuozhu Wen	Dalin Shi	2021-3
Optics			
Phytoplankton pigment absorption			
coefficient(a)	Xiaolong Yu	Kunshan Gao	2021-3
eventerent(a <sup>ph</sup> )			
Absorption coefficient of colored dissolved			
organic matter (a <sub>g</sub> )	Xiaolong Yu	Kunshan Gao	2021-3
5			
Remote sensing reflectance ( $R_{rs}$ )	Xiaolong Yu	Kunshan Gao	2021-3
PAR and R	Xiaolong Vu	Kunshan Gao	2021-3
rs	matrix I u	Transnan Oao	2021-2
Inherent optical profile	Xiaolong Yu	Kunshan Gao	2021-3

### Table 1 KK2003/GPpr15 Cruise Data Inventory

Aerosol thickness (AOT)	Xiaolong Yu	Kunshan Gao	2021-3
Biogeochemistry			
Macronutrients and low lever nutrients	Lifang Wang, Tao Huang	Minhan Dai	2021-3
DO, DIC, TA, pH	Ying Gao	Minhan Dai	2021-3
DOC	Peng Jiang	Minhan Dai	2021-3
DON, DOP	Peng Jiang	Xiaolin Li	2021-3
N <sub>2</sub> O	Wenbin Zou	Shuh-Ji Kao	2021-12
Chlorophyll a (Fluorescence)	Feipeng Xu	Bangqin Huang	2021-8
Phytoplankton pigment	Feipeng Xu	Bangqin Huang	2021-8
Primary production	Haoran Liu	Xin Liu	2021-8
Chl a (discrete samples)	Ze Chen	Dalin Shi	2021-3
FCM	Zuozhu Wen	Dalin Shi	2021-3
FCM(Macro omics)	Zuozhu Wen	Dalin Shi	2021-3
DNA/RNA	Zuozhu Wen	Dalin Shi	2021-10
Community Structure	Zuozhu Wen	Dalin Shi	2021-10
Functional gene analysis	Zuozhu Wen	Dalin Shi	2021-10
Biomarker	Wenkai Guan	Ruifeng Zhang	2021-6
Zooplankton multinet	Feipeng Xu	Bangqin Huang	2021-8
Bacterial respiration	Yuchen Zhang	Shaoling Shang	2021-8
Bacteria abundance	Yuchen Zhang	Shaoling Shang	2021-8
Bacterial diversity	Yuchen Zhang	Shaoling Shang	2021-8
On-deck Incubation			
FCM (on-deck incubation)	Ruotong Jiang	Dalin Shi	2021-3
Chl a (on-deck incubation)	Wenfang Lin	Dalin Shi	2021-3
Chl a (OA incubation)	Ze Chen	Dalin Shi	2021-3
DDN (on-deck incubation)	Hui Shen	Shuh-Ji Kao	2021-6
Nitrogen fixation rate (on-deck incubation)	Wenbin Zou	Shuh-Ji Kao	2021-6
Nitrogen fixation rate (+nutrients)	Zuozhu Wen	Dalin Shi	2021-10
Carbon fixation rate (+nutrients)	Zuozhu Wen	Dalin Shi	2021-10
DNA/RNA(+nutrients)	Zuozhu Wen	Dalin Shi	2021-10
N <sub>2</sub> O rate (on-deck incubation)	Hui Shen	Shuh-Ji Kao	2021-12
NP and RPP (on-deck incubation)	Hui Shen	Shuh-Ji Kao	2021-12
Surface Photosynthesis sequesters carbon	Guang Gao	Kunshan Gao	2021-8
Surface NPSi	Guang Gao	Kunshan Gao	2021-8
Surface community structure	Guang Gao	Kunshan Gao	2021-8
Rate			
Phytoplankton growth rate and	D ' II	<b>X</b> 7' <b>T</b> '	2021 0
microzooplankton grazing rate	Rui Huang	Xin Liu	2021-8
Nitrogen fixation rate	Ruotong Jiang, Zuozhu Wen	Dalin Shi	2021-10
Carbon fixation rate	Wenfang Lin, Zuozhu Wen	Dalin Shi	2021-10

Nitrogen fixation rate(underway)		Dalin Shi	2021-10
Carbon fixation rate(underway)		Dalin Shi	2021-10
Others			
Aerosol	Wenkai Guan	Jing Zhang	2021-12
Rain	Wanyang He	Jing Zhang	2021-12

### 3. Cruise Participants

Name	Email	Organization	<b>Shipboard Duties</b>	Subprojects
Xin Liu	liuxin1983@xmu.edu.cn	Xiamen University	Chief scientist	Subproject 3
Weifang	abany former adv an	Xiamen University	Chief scientist, clean CTD,	
Chenwi@xmu.edu.cn			in-situ pump, Th234	Subproject 4
Yongming	von om in abuon a Quenu adu on	Xiamen University		
Huang	yongminghuang@xmu.edu.cn		Chief scientist, FIA	Subproject 1
Wanyang	21100211024@sty ava adu an	Ocean University of		
He	21190311024@stu.ouc.edu.ch	China	Nd isotope	Subproject 1
Wenkai	awk1050460044@163.com	Ocean University of		
Guan	gwk1030409944@103.com	China	REEs et al.	Subproject 1
Zhan		Shanghai Jiaotong	TMC CTD Supertec,	
Shen	oucsnenxiaoznan@165.com	University	underway coordination	Subproject 1
Liping Ye	lpye@xmu.edu.cn	Xiamen University	TMC CTD Supertech	Subproject 1
Junbo	in the Original states of	Xiamen University	Dissolved and particulate	
Yang	Junbo@xmu.edu.cn		trace metal	Subproject 1
Nan	-1	Xiamen University		
Zhang	znnan@stu.xmu.edu.cn		Trace metal sampling	Subproject 1
Ziyuan		Shanghai Jiaotong		
Jiang	jiangziyuan@sjtu.edu.cn	University	Fe, Cd isotope	Subproject 1
Zhiyong		Xiamen University		
Cao	caozui@163.com		Turbulence	Subproject 1
Peng		Xiamen University		
Jiang	patrick@stu.xmu.edu.cn		DOC	Subproject 1
Lifang	1:6	Xiamen University		
Wang	mang@xmu.edu.cn		Macronutrients	Subproject 1
Tao	140	Xiamen University	Low level NH4, SUNA,	
Huang	nt@xmu.edu.cn		sampling	Subproject 1
Ruotong	240(71591)	Xiamen University	Nitrogen fixation rate,	
Jiang	2496/1381@qq.com		carbon fixation rate	Subproject 2
	sharr-200@1(2 a sur	Xiamen University	nifH gene abundance,	
Ze Chen	chenze300( <i>w</i> 163.com		activity	Subproject 2
Wenfang	1	Xiamen University	Metagenomic and	
Lin	lwi@xmu.edu.cn		transcriptome	Subproject 2
Zuozhu	1 2014 1	Xiamen University	Iron, phosphorus addition,	
Wen	wenzuozhu2014@stu.xmu.edu.cn		on-deck incubation	Subproject 2
		Xiamen University	High resolution surface	
Wenbin	zouwenbin465@xmu.edu.cn		nitrogen fixation rate and	
Zou	_		biological sample collection	Subproject 2
		Xiamen University	Nitrogen fixation DDN	
Hui Shen	snennui@stu.xmu.edu.cn		transmission culture at key	Subproject 2

Name	Email	Organization	<b>Shipboard Duties</b>	Subprojects
			stations	
Feipeng	vufainang@vmu adu an	Xiamen University		Subproject 3
Xu	xuteipeng@xittu.edu.en		Chla/Pigment/FCM	
Rui	huangrui0613@stu xmu edu en	Xiamen University		Subproject 3
Huang	nuangruioo15@stu.xinu.edu.en		Multinet	
Haoran	450783448@gg.com	Xiamen University		Subproject 3
Liu	430785448@qq.com		РР	
Yuchen	langyami@aa.com	Xiamen University	Bacteria production,	Subproject 3
Zhang	angyann@qq.com		bacteria respiration	
Xiaolong	xlvu@xmu edu cn	Xiamen University	Lw, Ed, c, bb, ap, ad, aph,	Subproject 3
Yu	xiy u@xinu.edu.en		ag, chla, PFT	
Guang	guang gao@xmu edu en	Xiamen University	Photosynthetic carbon	Subproject 3
Gao	guang.guo(@xinu.edu.en		fixation	
Yifan Ma	yifanma@stu.xmu.edu.cn	Xiamen University	POC, in-situ pump	Subproject 4
Xinting	raoxt@xmu.edu.cn	Xiamen University		Subproject 4
Rao	ruox(@xinu.edu.en		Si, in-situ pump	
Kan	zhangkan@ymu edu en	Xiamen University		Subproject 4
Zhang	zhangkan@xhiu.edu.eh		Particulate Fe, trap	
	gaoving@stu vmu edu en	Xiamen University	DO, carbonate, O2/Ar, TA,	Subproject 4
Ying Gao	gaoying@stu.xinu.edu.en		pH, pCO2 underway	
Xinru	15610092965@163.com	Xiamen University		
Zhang	13010072703/@103.com		230Th-232Th/210Po-210Pb	Subproject 4

#### Sampling and parameters 4.

#### 4.1. Physical oceanographic observations (Profs. Liu, and Lin)

#### 4.1.1. CTD Instrument information

Regular CTD profiles used the instrument of Seabird SBE 911 plus with serial numbers of 1181 and 1290. The instrument belongs to the R/V "Tan Kah Kee", with sensors include temperature, salinity, chlorophyll, dissolved oxygen, PAR, Suna, etc and attached to a rosette equipped with 24 Niskin 12L bottles. Compared with the regular CTD, trace metal clean CTD has no PAR and SUNA sensors, and other configurations are the same. Meanwhile, Acoustic Doppler Current Profiler (ADCP), Turbulence dissipation rate (VMP), Automatic Weather Station, underwater CTD, and Moving Vessel Profiler (MVP) were also applied.

Table 3 Instrument information				
Instrument	Parameters	<b>Station Number</b>	Cast Number	
<b>Regular CTD</b>	Temperature	18	172	
(SBE 911 plus)	Conductivity/Salinity			
	Pressure/Depth			
	Oxygen			
	Fluorescence			
	Beam Transmission			
	PAR/Irradiance			
				-





#### 4.1.2. Basic physical oceanographic background and turbulence characteristics

Sea Surface Temperature (SST) images in the July and August show the change of sea surface temperature during the cruise (**Figure 3**). The tropical / subtropical characteristics of the Western Pacific dominate the physical ocean background of the observation area. Stronger stratification in the upper ocean at 13°N transect but weaker stratification at 20°N transect (**Figure 4a, b**). Significant difference of salinity in vertical at 13°N while in horizontal at 20°N (**Figure 4c, d**). In this cruise, we have got direct measurements of micro scale shear from the free-fall vertical microstructure profiler (VMP-500) of 18 stations(64 casts) in the gyre. Further research needs to be carried out to reveal the underlying physical mechanism of weak mixing in this region.



Figure 3. Monthly mean Sea surface temperature from Himawari-8 (left: July, right: August)



Figure 4. Distribution of temperature and salinity(0-200m) along the 13°N&20°N transect References

de Boyer Montégut C, Madec G, Fischer A S, et al. Mixed layer depth over the global ocean: An examination of profile data and a profile-based climatology[J]. Journal of Geophysical Research: Oceans, 2004, 109(C12).

Kim S Y, Pak G, Lee H J, et al. Late-1980s Regime Shift in the Formation of the North Pacific Subtropical Mode Water[J]. Journal of Geophysical Research: Oceans, 2020, 125(2): e2019JC015700.

Whalen, C.B., J.A. MacKinnon & L.D. Talley (2018). Large-scale impacts of the mesoscale environment on mixing from wind-driven internal waves. Nature Geoscience, 11, 842–847.

#### 4.2. Apparent and Inherent Optical Properties (Prof. Shang)

#### 4.2.1. Remote Sensing of Reflectance

Remote sensing reflectance ( $R_{rs}(\lambda)$ , in sr<sup>-1</sup>), defined as the ratio of water-leaving radiance ( $L_w(\lambda)$ , in  $\mu$ W/cm<sup>2</sup>/nm/sr) to the downwelling irradiance just above the surface ( $E_d(0^+, \lambda)$ , in  $\mu$ W/cm<sup>2</sup>/nm), is a fundamental property in ocean color remote sensing (Mobley, 1994).

During the GPpr15 cruise,  $R_{rs}(\lambda)$  is determined by the in-water approach with a hyperspectral radiometer system (HyperPro II, Sea-Bird Scientific). HyperPro II was deployed to 80 ~ 150 m beneath the surface to measure the vertical profiles of the spectral upwelling radiance  $L_u(z, \lambda)$  with z the water depth. To determine  $L_w(\lambda)$ , spectral upwelling radiance just below the water surface  $(L_u(0-, \lambda))$  was first determined by extrapolating the profile measurement of  $L_u(z, \lambda)$  to the sea surface assuming an exponential decreasing. Then,  $L_u(0-, \lambda)$  can be converted to  $L_w(\lambda)$  following (Austin, 1974),

$$L_w(\lambda) = \frac{t}{n^2} L_u(0^-, \lambda), \qquad (0.1)$$

where *t* is the Fresnel transmittance of the air-sea interface ( $\sim 0.98$ ) and *n* is the refractive index of seawater ( $\sim 1.34$ ).

The incident surface irradiance  $E_s(\lambda)$  was concurrently recorded using a calibrated irradiance sensor (HyperOCR, Sea-Bird Scientific) mounted at the top of a pole on the deck, which is approximately 8 m above the sea surface to avoid any potential shadows. It is assumed that  $E_s(\lambda)$ is equivalent to the downwelling irradiance just above the surface, *i.e.*,  $E_s(\lambda) = E_d(0^+, \lambda)$ . Consequently,  $R_{rs}(\lambda)$  can be estimated as the ratio of  $L_w(\lambda)$  to  $E_s(\lambda)$ .

During the cruise, HyperPro II was deployed at 14 stations out of the total 21 stations. In the Mega Stations, such as WPS and M35, multiple deployments were carried out to investigate the daily variability of  $R_{rs}(\lambda)$ . In total, 20 deployments were carried out, and the sampling information is tabulated in Table 4.

Station	Date	Time (UTC+8h)
K2b	7/6/2020	11:00:27
KPR-1	7/7/2020	10:41:59
KPR-2	7/7/2020	15:42:02
WPS-1	7/14/2020	6:23:18
WPS-2	7/14/2020	11:46:40
WPS-3	7/15/2020	11:23:37
MR04	7/19/2020	12:23:06
MR05	7/21/2020	14:34:47
M35-1	7/24/2020	8:29:31
M35-2	7/25/2020	11:18:12
M35-3	7/26/2020	10:08:03
M32	7/27/2020	6:50:10
M22	8/2/2020	9:30:29
M20	8/4/2020	14:09:13
M18	8/5/2020	8:26:18
K8a-1	8/8/2020	9:51:38
K8a-2	8/9/2020	9:07:28
K9a	8/11/2020	11:26:40
K12a	8/15/2020	10:48:16
K13	8/17/2020	14:11:24

Table 4. Stations with valid  $R_{rs}$  measurements, along with dates/times.

For each deployment,  $3 \sim 4$  radiometric casts were carried out. The resultant  $R_{rs}(\lambda)$  is the average value of those casts after removing low-quality profile measurements. A total of 19valid  $R_{rs}(\lambda)$  were obtained.

#### **References:**

Mobley, C.D. (1994). Light and water: radiative transfer in natural waters. Academic Press. Austin, R.W. (1974). The remote sensing of spectral radiance from below the ocean surface. Optical aspects of oceanography, 317-344.

## 4.2.2. Photosynthetically Active Radiation (PAR) and the Diffuse Attenuation of Downwelling Irradiance $(K_d)$

The HyperPro II is also equipped with an irradiance sensor (HyperOCR, Sea-Bird Scientific) that can concurrently measure the spectral downwelling irradiance within the water column ( $E_d(z, \lambda)$ ). To account for the changes in the ambient light field during the cast, the measured  $E_d(z, \lambda)$  was normalized by the deck cell-measured surface irradiance (Fargion and Mueller, 2000),

$$E'_{d}(z,\lambda) = \frac{E_{d}(z,\lambda) \times E_{s}(t(0^{-}),\lambda)}{E_{s}(t(z),\lambda)},$$
(0.2)

where  $E_s(t(z), \lambda)$  is the surface irradiance measured on deck at the time t(z) when the radiometer

was at depth z, and  $E_s(t(0^-), \lambda)$  is the measurement at time  $t(0^-)$  when the radiometer was at the sea surface.

The normalized  $E'_d(z,\lambda)$  is later used to calculate the photosynthetically active radiation at

different depths (PAR(z), in  $\mu W/cm^2$ ) by integrating  $E'_d(z, \lambda)$  between 400 and 700 nm.

The diffuse attenuation of downwelling irradiance,  $K_d(\lambda)$  (in m<sup>-1</sup>), can be calculated as the slope of the linear regression between  $\ln(E'_d(z,\lambda))$  and the depth z.

Sampling locations are the same as those measurements of  $R_{rs}(\lambda)$  in Section 4.2.1.

The estimated PAR(z) from  $E'_{d}(z,\lambda)$  is plotted against the depth z, with the vertical profiles of PAR(z) for each valid cast.

#### **References:**

Fargion, G.S., & Mueller, J.L. (2000). Ocean optics protocols for satellite ocean color sensor validation, Revision 2. National Aeronautics and Space Administration, Goddard Space Flight Center.

#### 4.2.3. Attenuation, absorption, and backscattering coefficients

A cage with multiple IOPs instruments, including the WetLabs ac-s and bb9 (Sea-Bird Scientific), was deployed to a maximum depth of 250 m to measure the inherent optical properties of the water column. Specifically, WetLab ac-s determines the spectral attenuation coefficient ( $c(\lambda)$ , in m<sup>-1</sup>) and spectral absorption coefficient for non-water component ( $a(\lambda)$ , in m<sup>-1</sup>), *i.e.*, the phytoplankton, colored dissolved organic matter, and the non-algal particles. ac-s takes measurements from 400 to 750 nm with a spectral resolution of ~ 4 nm. bb9 has nine spectral channels. Seven of those channels are used to determine the total backscattering coefficient ( $b_b(\lambda)$ , in m<sup>-1</sup>) at 412, 440, 488, 532, 595, 695, and 715 nm, respectively. The rest two channels are used to their concentrations, respectively.

The determined spectral a and c by ac-s were corrected for the salinity and temperature effects (Pegau et al., 1997), which was implemented by the Compass Host Software (Sea-Bird Scientific) during post-processing. The measured absorption coefficient was further corrected for the scattering effects, according to Röttgers et al. (2013).

The IOPs cage was deployed 11 times at mainly the Mega stations, such as K2b, WPS, and M35. Three additional casts were carried out at stations M30, K8a, and K13, respectively. The sampling information, including the location and date/time, of each cast are provided in Table 5.

Station	Date	Time (UTC+8h)
K2b	7/6/2020	7:16
K2b	7/6/2020	13:20
K2b	7/7/2020	17:23
WPS	7/13/2020	23:37
WPS	7/14/2020	13:45
WPS	7/15/2020	14:39
M35	7/24/2020	8:57
M35	7/25/2020	13:16
M30	7/28/2020	9:10
K8a	8/8/2020	10:54
K13	8/17/2020	15:00

Table 5. Stations with valid IOPs casts, along with dates/times.

#### **References:**

Pegau, W.S., Gray, D., & Zaneveld, J.R.V. (1997). Absorption and attenuation of visible and near-infrared light in water: dependence on temperature and salinity. Applied Optics, 36, 6035-6046.

Röttgers, R., McKee, D., & Woźniak, S.B. (2013). Evaluation of scatter corrections for ac-9 absorption measurements in coastal waters. Methods in Oceanography, 7, 21-39

#### 4.2.4. Component Absorption Coefficients

Discrete water samples were collected using Niskin bottles at the site of the in-water optical measurements at eight depths from just beneath the surface (~ 5 m) to a maximum of 200 m. The depth increments for each cast were set primary as 5m, 15m, 25m, 50m, 75m, 100m, 150m, and 200m, respectively. However, for each cast, one of these eight depths, whichever close to the depth of the chlorophyll maximum (DCM), was replaced by water collected from DCM. Preparation of the absorption samples for CDOM and particles on shipboard were conformed to the NASA protocols (Mueller et al., 2003).

Briefly, for the particle absorption samples, 5000 mL seawater was gravity-filtered onto GF/F glass microfiber filters (Whatman  $\mathbb{R}$ , pore size ~ 0.7 µm) under a low vacuum immediately after the sampling. The particle-loaded filters were later stored in the liquid nitrogen during the cruise. Reference filters were also prepared every day following the same steps as the samples, except that only 200 mL Mill-Q waters were used instead of the seawater. For CDOM absorption samples, 200 mL seawater was gravity-filtered using a 0.22 µm polycarbonate membrane (Millipore) under a low vacuum immediately after the sampling. The membranes were soaked in 10% HCL for 15 min and then rinsed with Milli-Q water three times before the filtration. The filtered CDOM samples were collected in borosilicate glass vials and then stored in the -20 °C refrigerator during the cruise.

A dual-beam spectrophotometer (PerkinElmer Lambda 850) was used in the laboratory to determine the absorption of CDOM samples, where CDOM absorption coefficient ( $a_g(\lambda)$ , in m<sup>-1</sup>) were measured from 250 to 900 nm using a 10 cm quartz cuvette referenced to the Milli-Q water. The same spectrophotometer with the integrating-sphere attachment was used to determine the particulate absorption ( $a_p(\lambda)$ , in m<sup>-1</sup>) for the spectral domain between 350 and 800 nm using the quantitative filter approach (QFT) (Tassan and Ferrari, 1995). A blank GF/F filter was used as the reference after filtering 500 mL Mill-Q waters onto the filter. The particle-loaded filters, as well as the reference filters, were later bleached using methanol for half an hour and then rinsed with Milli-Q water three times. The bleached filters were then used to determine the absorption coefficients of non-algal particles ( $a_{nap}(\lambda)$ , in m<sup>-1</sup>) following the same procedures as that for  $a_p(\lambda)$ . The absorption coefficients of phytoplankton ( $a_{ph}(\lambda)$ , in m<sup>-1</sup>) are calculated as the difference between  $a_p(\lambda)$  and  $a_{nap}(\lambda)$ .

#### **References:**

Mueller, J.L., Fargion, G.S., McClain, C.R., Pegau, S., Zaneveld, J.R.V., Mitchell, B.G., Kahru, M., Wieland, J., & Stramska, M. (2003). Ocean optics protocols for satellite ocean color sensor validation, revision 4, volume IV: Inherent optical properties: Instruments, characterizations, field measurements and data analysis protocols. NASA Tech. Memo, 01674-01670.

Tassan, S., & Ferrari, G.M. (1995). An alternative approach to absorption measurements of aquatic particles retained on filters. Limnology and Oceanography, 40, 1358-1368.

#### 4.2.5. Secchi Disk Depth

A white circular disk, 30 cm in diameter, was used to measure the Secchi disk depth ( $Z_{sd}$ , in m). Operationally, the disk was lowered into the water until it can no longer be seen by the observer. The depth of disappearance is recorded as  $Z_{sd}$ . The same procedure was repeated three times, with three readings that were later averaged as the final  $Z_{sd}$ . The deployment of the Secchi disk is strongly affected by both ocean currents and wind speeds, which could result in a large angle of drift relative to the normal. Therefore, those deployments with the angle of drift larger than ~ 30° were considered invalid observations. Secchi disk was only deployed at some of the stations depending on the sea state and wind conditions.

#### 4.2.6. Aerosol Optical Thickness

Aerosol optical thickness (AOT) was measured by a Microtops II Sunphotometer (Model 540, Solar Light), which provides AOT measurements at five channels centered at 380, 500, 675, 870, and 936 nm, respectively. To measure AOT, the Sunophtometer was pointed towards the sun, and its position was manually adjusted so that the image of the sun appears in the "Sun target" window and centered on the bull's-eye. Measurements of AOT were only carried out under clear conditions, with no cloud or haze covering the sun. At each sampling site, continuous ten scans were made, which were later averaged to compute the AOT for the respective sites. A total of 81 valid AOT measurements were carried out.

#### 4.3. Chemical Oceanography Trace Metal Clean CTD (Profs. Cai, Dai and Zhang)

#### 4.3.1. Instrument information

The Clean CTD Sampling System includes clean sampling bottles (OTE C-FREE or GO Niskin-X), a SBE 911plus CTD attached to a 32G-24P sampling carousel, a clean sampling van, a clean analysis van, and a clean CTD winch with electrical control container. Table 6 shows the detailed information of the clean CTD sampling system.

Instrument	Specification	Manufacturer
Clean someling Dattle	C-FREE 114, 12L	Ocean Test Equipment
Clean sampling Bottle	Niskin-X, 12L	General Oceanics
CTD and sampling carousel	CTD: SBE 911Plus; Carousel: 32G-24P, 24-position for use with 12-liter sampling bottles, Integrated with 2 sets of temperature-conductivity sensors, SBE43 dissolved oxygen sensor, WET Labs C-Star, WET Labs ECO-FLrtd, and VA-500 altimeter	SeaBird
Clean CTD winch	2550*2350*1950mm; contain 8000m Vectran conducting cable	Kleyfrance
Winch electrical control container	3000*2450*2850mm; Contain portable control console	Kleyfrance
Sampling van	Custom, 606*244*259cm, Class1000	GeOceanTech Co., Ltd.
Analysis van	Custom, 606*244*259cm, Class1000	GeOceanTech Co., Ltd.

#### **Table 6 Instrument information**

#### 4.3.2. Parameters

Parameters obtained with Clean CTD sampling system include nutrient concentrations and trace element concentrations and isotopic compositions:

- 1) Ca and Nutrient concentrations: NO<sub>3</sub><sup>-</sup>, NO<sub>2</sub><sup>-</sup>, NH<sub>4</sub><sup>+</sup>, PO<sub>4</sub><sup>3-</sup>, SiO<sub>3</sub><sup>2-</sup>
- 2) Trace metal concentrations: REE, Fe (FIA), Fe (ICP-MS), Cu, Zn, Cd, Ni, Pb, Co, Mn (FIA), Mn (ICP-MS), Al, THg, MeHg, Mo, V
- 3) Trace metal isotopes:  $\delta^{56/54}$ Fe,  $\delta^{66/64}$ Zn,  $\delta^{114/110}$ Cd,  $^{206/207}$ Pb,  $^{208/207}$ Pb,  $^{206/204}$ Pb,  $\delta^{65}$ Cu

#### 4.3.3. Objectives

To investigate the distribution of TEIs in the wNP and to improve the understanding of biogeochemical cycles of TEIs.

#### 4.3.4. Analytical Methods

#### 1) Ca and Nutrients

Samples for  $Ca^{2+}$  analysis were stored in 125 ml acid-cleaned polyethylene bottles with Parafilm wrapped around the cap. In the laboratory,  $Ca^{2+}$  was determined using the classic ethylene glycol tetraacetic acid (EGTA) titration method and Calcium-Ion Selective Electrode for end-point detection.

Nutrient concentrations will be analyzed onboard with a Four-channel Continuous Flow Technicon AA3 Auto-Analyzer (Bran-Lube, GmbH) according to classic colorimetric methods. Nanomolar level  $NO_3^-$  is determined on a continuous flow analysis system combined with a liquid waveguide capillary flow cell. SRP at nanomolar level is determined using a flow injection system onboard within 1 day of sampling. Detailed information was described in section **4.1.6**.

#### 2) Trace metal concentration and their isotopes

The dissolved Fe (FIA) concentration will be analyzed following a modified flow injection-based luminol chemiluminescence method with a detection limit of 0.020 nM. Fe(III) in seawater samples will be reduced to Fe(II) by sodium sulfite before preconcentration. A mini column packed with Nobias Chelate-PA1 resin (Hitachi) will be used for on-line preconcentration of iron from seawater matrix at pH  $6.2\pm0.1$ . After preconcentration, iron will be eluted from the Nobias Chelate-PA1 column by 0.08 mol/L HCl solution and then merged with luminol solution at a reaction pH of  $10.1\pm0.1$  to produce chemiluminescence for the detection.

The concentrations of dissolved Al(FIA) and Mn(FIA) will be analyzed by the flow injection analysis methods with on-line preconcentration following the protocol described in Brown and Bruland (2008) and Aguilar-Islas and Bruland (2006). The detection limits are 0.18 nM and 0.2 nM for Al and Mn, respectively. The concentrations of dissolved REE, Fe (ICP-MS), Cu, Zn, Cd, Ni, Pb, Co, Mo, V will be pre-concentrated and analyzed by inductively coupled plasma mass spectrometry (ICP-MS, Thermo ICAP-Q/Element XR) using isotopic dilution or standard addition method.

For the total Hg concentration analysis, samples will be oxidized with 0.05% bromine monochloride (BrCl) solution or equivalent for at least 1 hour. Then excess halogens will be removed by mixing with 0.05% v/v hydroxylamine hydrochloride (NH<sub>2</sub>OH•HCl) solution for at least 5 minutes, and final reduction will be conducted with 0.05% v/v stannous chloride (SnCl<sub>2</sub>) solution. Hg<sup>0</sup> in solution will be purged and trapped on gold or gold-coated sand (or the equivalent). Purging should proceed at a volumetric flow rate of no more than 1 L min<sup>-1</sup> (we recommend 0.5 L min<sup>-1</sup>) until a volume of gas of at least 15 times the volume of liquid has been

sparged. For MeHg analysis, HCl (conc.) is added to 250 ml seawater sample to form a final concentration of 0.5% and the samples are stored at -20 °C before analysis. Upon arrival at the laboratory, 250 ml seawater sample is digested at 2% H<sub>2</sub>SO<sub>4</sub> for > 12h. After that, the sample is first neutralized with 7.5 mL of 50% KOH, and then buffered to pH=5 with 3.75 mL of 2 M Na-Acetate/Acetic buffer, the pH should be checked and adjusted as necessary with small additions of strong acid (H<sub>2</sub>SO<sub>4</sub>) or strong base (KOH). 0.18 mL of 1% NaTEB will be added to the buffered 250 ml sample, allowing each sample to react for at least 15 minutes, and then sparging the formed MeHgEt from the sample to a Tenax trap. MeHgEt on the trap will then be detected by a packed column GC (OV-3)-AFS.

 $\delta^{56/54}$ Fe,  $\delta^{66/64}$ Zn,  $\delta^{114/110}$ Cd will be analyzed using double spike method followed by the batch resin extraction. <sup>206/207</sup>Pb, <sup>208/207</sup>Pb, <sup>206/204</sup>Pb will be purified using column chemistry method.  $\delta^{65}$ Cu will be analyzed using standard bracketing method after batch resin extraction. All of these isotopes will be measured on a multiple collector ICP-MS (Conway et al., 2013).

#### 4.3.5. Sampling Information

Samples for trace elements were collected by 12-liter Niskin-X bottles mounted onto a clean rosette sampling assembly, equipped with a CTD recorder (Sea-Bird SBE911Plus, see 4.2.1 for detailed information). During this cruise we collected 368 layer seawater samples at 10 clean stations and 5 mega stations. 160 surface water samples were collected by a homemade underway fish-towing system.

Unfiltered and filtered seawater samples were subsampled for each depth. Detailed sampling information is given in Table 7. To obtain filtered samples, seawater was filtered in-line from Niskin-X bottles in the clean van through AcroPak 1000 cartridges (pore size of 0.8/0.45 mm) into pre-cleaned LDPE/HDPE bottles. The filtrate was acidified to a pH of ~2 with ultra-clean 6 N HCl (Fisher Scientific optima grade) in a 100-class clean van onboard and stored for more than one month before analysis.

	Dattla				Unfiltere	ed				Filte	red	
Station	Dottie	Depth	0- 0 0-1		Low		<b>N</b> 4 • 4	DEE	A 1/3/K	Fe	Fe/Zn/Cd	TEIs
	Number		Ca&Sai	AI	N&P	I EIS	Nutrient	REES	Al/Mn	(FIA)	isotopes	(ICP-MS)
K2b	1	500	0.125	0.25		0.25	0.125	0.5	0.25	0.125	2	4.5
K2b	2	400	0.125	0.25		0.25	0.125	0.5	0.25	0.125	2	4.5
K2b	3	300	0.125	0.25		0.25	0.125	0.5	0.25	0.125	2	4.5
K2b	4	250	0.125	0.25		0.25	0.125	0.5	0.25	0.125	2	4.5
K2b	5	200	0.125	0.25		0.25	0.125	0.5	0.25	0.125	2	4.5
K2b	6	190	0.125	0.25		0.25	0.125	0.5	0.25	0.125	2	4.5
K2b	7	180	0.125	0.25		0.25	0.125	0.5	0.25	0.125	2	4.5
K2b	8	170	0.125	0.25		0.25	0.125	0.5	0.25	0.125	2	4.5

Table 7 Sampling information for each TEIs clean cast

K2b	9	160	0.125	0.25		0.25	0.125	0.5	0.25	0.125	2	4.5
K2b	10	150	0.125	0.25	0.25	0.25	0.125	0.5	0.25	0.125	2	4.5
K2b	11	140	0.125	0.25	0.25	0.25	0.125	0.5	0.25	0.125	2	4.5
K2b	12	130	0.125	0.25	0.25	0.25	0.125	0.5	0.25	0.125	2	4.5
K2b	13	120	0.125	0.25	0.25	0.25	0.125	0.5	0.25	0.125	2	4.5
K2b	14	110	0.125	0.25	0.25	0.25	0.125	0.5	0.25	0.125	2	4.5
K2b	15	100	0.125	0.25	0.25	0.25	0.125	0.5	0.25	0.125	2	4.5
K2b	16	90	0.125	0.25	0.25	0.25	0.125	0.5	0.25	0.125	2	4.5
K2b	17	80	0.125	0.25	0.25	0.25	0.125	0.5	0.25	0.125	2	4.5
K2b	18	70	0.125	0.25	0.25	0.25	0.125	0.5	0.25	0.125	2	4.5
K2b	19	60										
K2b	20	50	0.125	0.25	0.25	0.25	0.125	0.5	0.25	0.125	2	4.5
K2b	21	40	0.125	0.25	0.25	0.25	0.125	0.5	0.25	0.125	2	4.5
K2b	22	30	0.125	0.25	0.25	0.25	0.125	0.5	0.25	2.125	2	4.5
K2b	23	20	0.125	0.25	0.25	0.25	0.125	0.5	0.25	0.125	2	4.5
K2b	24	10	0.125	0.25	0.25	0.25	0.125	0.5	0.25	0.125	2	4.5
K2b	1	5200	0.125	0.25		0.25	0.125	0.5	0.25	0.125	2	4.5
K2b	2	4500	0.125	0.25		0.25	0.125	0.5	0.25	0.125	2	4.5
K2b	3	4000	0.125	0.25		0.25	0.125	0.5	0.25	0.125	2	4.5
K2b	4	3600	0.125	0.25		0.25	0.125	0.5	0.25	0.125	2	4.5
K2b	5	3200	0.125	0.25		0.25	0.125	0.5	0.25	0.125	2	4.5
K2b	6	3000	0.125	0.25		0.25	0.125	0.5	0.25	0.125	2	4.5
K2b	7	2600	0.125	0.25		0.25	0.125	0.5	0.25	0.125	2	4.5
K2b	8	2200	0.125	0.25		0.25	0.125	0.5	0.25	0.125	2	4.5
K2b	9	2000	0.125	0.25		0.25	0.125	0.5	0.25	0.125	2	4.5
K2b	10	1750	0.125	0.25		0.25	0.125	0.5	0.25	0.125	2	4.5
K2b	11	1600	0.125	0.25		0.25	0.125	0.5	0.25	0.125	2	4.5
K2b	12	1400	0.125	0.25		0.25	0.125	0.5	0.25	0.125	2	4.5
K2b	13	1200	0.125	0.25		0.25	0.125	0.5	0.25	0.125	2	4.5
K2b	14	1000	0.125	0.25		0.25	0.125	0.5	0.25	0.125	2	4.5
K2b	15	900	0.125	0.25		0.25	0.125	0.5	0.25	0.125	2	4.5
K2b	16	800	0.125	0.25		0.25	0.125	0.5	0.25	0.125	2	4.5
K2b	17	700	0.125	0.25		0.25	0.125	0.5	0.25	0.125	2	4.5
K2b	18	600	0.125	0.25		0.25	0.125	0.5	0.25	0.125	2	4.5
KPR	11	1000	0.125	0.25		0.25	0.125	0.5	0.25	0.125	2	4.5
KPR	12	800	0.125	0.25		0.25	0.125	0.5	0.25	0.125	2	4.5
KPR	13	600	0.125	0.25		0.25	0.125	0.5	0.25	0.125	2	4.5
KPR	14	500	0.125	0.25		0.25	0.125	0.5	0.25	0.125	2	4.5
KPR	15	400	0.125	0.25		0.25	0.125	0.5	0.25	0.125	2	4.5
KPR	16	300	0.125	0.25		0.25	0.125	0.5	0.25	0.125	2	4.5
KPR	17	200	0.125	0.25		0.25	0.125	0.5	0.25	0.125	2	4.5
KPR	18	155	0.125	0.25	0.25	0.25	0.125	0.5	0.25	0.125	2	4.5
KPR	19	125	0.125	0.25	0.25	0.25	0.125	0.5	0.25	0.125	2	4.5

KPR	20	100	0.125	0.25	0.25	0.25	0.125	0.5	0.25	0.125	2	4.5
KPR	21	75	0.125	0.25	0.25	0.25	0.125	0.5	0.25	0.125	2	4.5
KPR	22	50	0.125	0.25	0.25	0.25	0.125	0.5	0.25	2.125	2	4.5
KPR	23	25	0.125	0.25	0.25	0.25	0.125	0.5	0.25	0.125	2	4.5
KPR	24	10	0.125	0.25	0.25	0.25	0.125	0.5	0.25	0.125	2	4.5
WPS	1	500	0.125	0.25		0.25	0.125	0.5	0.25	0.125	2	4.5
WPS	2	400	0.125	0.25		0.25	0.125	0.5	0.25	0.125	2	4.5
WPS	3	300	0.125	0.25		0.25	0.125	0.5	0.25	0.125	2	4.5
WPS	4	250	0.125	0.25		0.25	0.125	0.5	0.25	0.125	2	4.5
WPS	5	200	0.125	0.25		0.25	0.125	0.5	0.25	0.125	2	4.5
WPS	6	190	0.125	0.25		0.25	0.125	0.5	0.25	0.125	2	4.5
WPS	7	180	0.125	0.25		0.25	0.125	0.5	0.25	0.125	2	4.5
WPS	8	170	0.125	0.25		0.25	0.125	0.5	0.25	0.125	2	4.5
WPS	9	160	0.125	0.25		0.25	0.125	0.5	0.25	0.125	2	4.5
WPS	10	150	0.125	0.25	0.25	0.25	0.125	0.5	0.25	0.125	2	4.5
WPS	11	140	0.125	0.25	0.25	0.25	0.125	0.5	0.25	0.125	2	4.5
WPS	12	130	0.125	0.25	0.25	0.25	0.125	0.5	0.25	0.125	2	4.5
WPS	13	120	0.125	0.25	0.25	0.25	0.125	0.5	0.25	0.125	2	4.5
WPS	14	110	0.125	0.25	0.25	0.25	0.125	0.5	0.25	0.125	2	4.5
WPS	15	100	0.125	0.25	0.25	0.25	0.125	0.5	0.25	0.125	2	4.5
WPS	16	90	0.125	0.25	0.25	0.25	0.125	0.5	0.25	0.125	2	4.5
WPS	17	80	0.125	0.25	0.25	0.25	0.125	0.5	0.25	0.125	2	4.5
WPS	18	70	0.125	0.25	0.25	0.25	0.125	0.5	0.25	0.125	2	4.5
WPS	19	60	0.125	0.25	0.25	0.25	0.125	0.5	0.25	0.125	2	4.5
WPS	20	50	0.125	0.25	0.25	0.25	0.125	0.5	0.25	0.125	2	4.5
WPS	21	40	0.125	0.25	0.25	0.25	0.125	0.5	0.25	0.125	2	4.5
WPS	22	30	0.125	0.25	0.25	0.25	0.125	0.5	0.25	2.125	2	4.5
WPS	23	20	0.125	0.25	0.25	0.25	0.125	0.5	0.25	0.125	2	4.5
WPS	24	10	0.125	0.25	0.25	0.25	0.125	0.5	0.25	0.125	2	4.5
WPS	1	4500	0.125	0.25		0.25	0.125	0.5	0.25	0.125	2	4.5
WPS	2	4200	0.125	0.25		0.25	0.125	0.5	0.25	0.125	2	4.5
WPS	3	4000	0.125	0.25		0.25	0.125	0.5	0.25	0.125	2	4.5
WPS	4	3800	0.125	0.25		0.25	0.125	0.5	0.25	0.125	2	4.5
WPS	5	3400	0.125	0.25		0.25	0.125	0.5	0.25	0.125	2	4.5
WPS	6	3000	0.125	0.25		0.25	0.125	0.5	0.25	0.125	2	4.5
WPS	7	2800	0.125	0.25		0.25	0.125	0.5	0.25	0.125	2	4.5
WPS	8	2400	0.125	0.25		0.25	0.125	0.5	0.25	0.125	2	4.5
WPS	9	2000	0.125	0.25		0.25	0.125	0.5	0.25	0.125	2	4.5
WPS	10	1800	0.125	0.25		0.25	0.125	0.5	0.25	0.125	2	4.5
WPS	11	1600	0.125	0.25		0.25	0.125	0.5	0.25	0.125	2	4.5
WPS	12	1370	0.125	0.25		0.25	0.125	0.5	0.25	0.125	2	4.5
WPS	13	1200	0.125	0.25		0.25	0.125	0.5	0.25	0.125	2	4.5
1		1000	0 105	0.25		0.25	0.125	0.5	0.25	0.125	r	4.5

	WPS	15	900	0.125	0.25		0.25	0.125	0.5	0.25	0.125	2	4.5
	WPS	16	800	0.125	0.25		0.25	0.125	0.5	0.25	0.125	2	4.5
	WPS	17	700	0.125	0.25		0.25	0.125	0.5	0.25	0.125	2	4.5
	WPS	18	600	0.125	0.25		0.25	0.125	0.5	0.25	0.125	2	4.5
Ī	MR04	1	2700	0.125			0.25	0.125	0.5	0.25	0.125	2	4.5
	MR04	2	2500	0.125			0.25	0.125	0.5	0.25	0.125	2	4.5
	MR04	3	2000	0.125			0.25	0.125	0.5	0.25	0.125	2	4.5
	MR04	4	1500	0.125			0.25	0.125	0.5	0.25	0.125	2	4.5
	MR04	5	1200	0.125			0.25	0.125	0.5	0.25	0.125	2	4.5
	MR04	6	1000	0.125			0.25	0.125	0.5	0.25	0.125	2	4.5
	MR04	7	900	0.125			0.25	0.125	0.5	0.25	0.125	2	4.5
	MR04	8	800	0.125			0.25	0.125	0.5	0.25	0.125	2	4.5
	MR04	9	700	0.125			0.25	0.125	0.5	0.25	0.125	2	4.5
	MR04	10	600	0.125			0.25	0.125	0.5	0.25	0.125	2	4.5
	MR04	11	500	0.125			0.25	0.125	0.5	0.25	0.125	2	4.5
	MR04	12	400	0.125			0.25	0.125	0.5	0.25	0.125	2	4.5
	MR04	13	300	0.125			0.25	0.125	0.5	0.25	0.125	2	4.5
	MR04	14	200	0.125			0.25	0.125	0.5	0.25	0.125	2	4.5
	MR04	15	180	0.125			0.25	0.125	0.5	0.25	0.125	2	4.5
	MR04	16	165	0.125			0.25	0.125	0.5	0.25	0.125	2	4.5
	MR04	17	150	0.125		0.25	0.25	0.125	0.5	0.25	0.125	2	4.5
	MR04	18	125	0.125		0.25	0.25	0.125	0.5	0.25	0.125	2	4.5
	MR04	19	100	0.125		0.25	0.25	0.125	0.5	0.25	0.125	2	4.5
	MR04	20	80	0.125		0.25	0.25	0.125	0.5	0.25	0.125	2	4.5
	MR04	21	60	0.125		0.25	0.25	0.125	0.5	0.25	0.125	2	4.5
	MR04	22	40	0.125		0.25	0.25	0.125	0.5	0.25	2.125	2	4.5
	MR04	23	20	0.125		0.25	0.25	0.125	0.5	0.25	0.125	2	4.5
	MR04	24	10	0.125		0.25	0.25	0.125	0.5	0.25	0.125	2	4.5
	MR05	1	1000	0.125			0.25	0.125	0.5	0.25	0.125	2	4.5
	MR05	2	800	0.125			0.25	0.125	0.5	0.25	0.125	2	4.5
	MR05	3	500	0.125			0.25	0.125	0.5	0.25	0.125	2	4.5
	MR05	4	300	0.125			0.25	0.125	0.5	0.25	0.125	2	4.5
	MR05	5	200	0.125			0.25	0.125	0.5	0.25	0.125	2	4.5
	MR05	6	175	0.125			0.25	0.125	0.5	0.25	0.125	2	4.5
	MR05	7	150	0.125			0.25	0.125	0.5	0.25	0.125	2	4.5
	MR05	8	120	0.125			0.25	0.125	0.5	0.25	0.125	2	4.5
	MR05	9	110	0.125			0.25	0.125	0.5	0.25	0.125	2	4.5
	MR05	10	100	0.125			0.25	0.125	0.5	0.25	0.125	2	4
	MR05	11	75	0.125			0.25	0.125	0.5	0.25	0.125	2	4.5
	MR05	22	50	0.125			0.25	0.125	0.5	0.25	2.125	2	4.5
	MR05	23	25	0.125			0.25	0.125	0.5	0.25	0.125	2	4.5
	MR05	24	10	0.125			0.25	0.125	0.5	0.25	0.125	2	4.5
	M35	1	500	0.125			0.25	0.125	0.5	0.25	0.125	2	4.5

M35	2	400	0.125		0.25	0.125	0.5	0.25	0.125	2	4.5
M35	3	300	0.125		0.25	0.125	0.5	0.25	0.125	2	4.5
M35	4	250	0.125		0.25	0.125	0.5	0.25	0.125	2	4.5
M35	5	200	0.125		0.25	0.125	0.5	0.25	0.125	2	4.5
M35	6	190	0.125		0.25	0.125	0.5	0.25	0.125	2	4.5
M35	7	180	0.125		0.25	0.125	0.5	0.25	0.125	2	4.5
M35	8	170	0.125		0.25	0.125	0.5	0.25	0.125	2	4.5
M35	9	160	0.125		0.25	0.125	0.5	0.25	0.125	2	4.5
M35	10	150	0.125	0.25	0.25	0.125	0.5	0.25	0.125	2	4.5
M35	11	140	0.125	0.25	0.25	0.125	0.5	0.25	0.125	2	4.5
M35	12	130	0.125	0.25	0.25	0.125	0.5	0.25	0.125	2	4.5
M35	13	120	0.125	0.25	0.25	0.125	0.5	0.25	0.125	2	4.5
M35	14	110	0.125	0.25	0.25	0.125	0.5	0.25	0.125	2	4.5
M35	15	100	0.125	0.25	0.25	0.125	0.5	0.25	0.125	2	4.5
M35	16	90	0.125	0.25	0.25	0.125	0.5	0.25	0.125	2	4.5
M35	17	80	0.125	0.25	0.25	0.125	0.5	0.25	0.125	2	4.5
M35	18	70	0.125	0.25	0.25	0.125	0.5	0.25	0.125	2	4.5
M35	19	60	0.125	0.25	0.25	0.125	0.5	0.25	0.125	2	4.5
M35	20	50	0.125	0.25	0.25	0.125	0.5	0.25	0.125	2	4.5
M35	21	40	0.125	0.25	0.25	0.125	0.5	0.25	0.125	2	4.5
M35	22	30	0.125	0.25	0.25	0.125	0.5	0.25	2.125	2	4.5
M35	23	20	0.125	0.25	0.25	0.125	0.5	0.25	0.125	2	4.5
M35	24	10	0.125	0.25	0.25	0.125	0.5	0.25	0.125	2	4.5
M35	1	5150	0.125		0.25	0.125	0.5	0.25	0.125	2	4.5
M35	2	5000	0.125		0.25	0.125	0.5	0.25	0.125	2	4.5
M35	3	4800	0.125		0.25	0.125	0.5	0.25	0.125	2	4.5
M35	4	4500	0.125		0.25	0.125	0.5	0.25	0.125	2	4.5
M35	5	4000									
M35	6	3500	0.125		0.25	0.125	0.5	0.25	0.125	2	4.5
M35	7	3000	0.125		0.25	0.125	0.5	0.25	0.125	2	4.5
M35	8	2500	0.125		0.25	0.125	0.5	0.25	0.125	2	4.5
M35	9	2000	0.125		0.25	0.125	0.5	0.25	0.125	2	4.5
M35	10	1800	0.125		0.25	0.125	0.5	0.25	0.125	2	4.5
M35	11	1600	0.125		0.25	0.125	0.5	0.25	0.125	2	4.5
M35	12	1400	0.125		0.25	0.125	0.5	0.25	0.125	2	4
M35	13	1200	0.125		0.25	0.125	0.5	0.25	0.125	2	4.5
M35	14	1000	0.125		0.25	0.125	0.5	0.25	0.125	2	4.5
M35	15	900	0.125		0.25	0.125	0.5	0.25	0.125	2	4.5
M35	16	800	0.125		0.25	0.125	0.5	0.25	0.125	2	4.5
M35	17	700	0.125		0.25	0.125	0.5	0.25	0.125	2	4.5
M35	18	600	0.125		0.25	0.125	0.5	0.25	0.125	2	4
M35	19	500	0.125		0.25	0.125	0.5	0.25	0.125	2	4.5
M30	1	5700	0.125		0.25	0.125	0.5	0.25	0.125	2	4.5

M30	2	5000	0.125		0.25	0.125	0.5	0.25	0.125	2	4.5
M30	3	4500	0.125		0.25	0.125	0.5	0.25	0.125	2	4.5
M30	4	4000	0.125		0.25	0.125	0.5	0.25	0.125	2	4
M30	5	3500	0.125		0.25	0.125	0.5	0.25	0.125	2	3
M30	6	3000	0.125		0.25	0.125	0.5	0.25	0.125	2	4.5
M30	7	2500	0.125		0.25	0.125	0.5	0.25	0.125	2	4.5
M30	8	2000	0.125		0.25	0.125	0.5	0.25	0.125	2	4.5
M30	9	1500	0.125		0.25	0.125	0.5	0.25	0.125	2	4.5
M30	10	1250	0.125		0.25	0.125	0.5	0.25	0.125	2	4.5
M30	11	1000	0.125		0.25	0.125	0.5	0.25	0.125	2	4.5
M30	12	800	0.125		0.25	0.125	0.5	0.25	0.125	2	4.5
M30	13	500	0.125		0.25	0.125	0.5	0.25	0.125	2	4.5
M30	14	400	0.125		0.25	0.125	0.5	0.25	0.125	2	4.5
M30	15	300	0.125		0.25	0.125	0.5	0.25	0.125	2	4.5
M30	16	200	0.125		0.25	0.125	0.5	0.25	0.125	2	4.5
M30	17	175	0.125		0.25	0.125	0.5	0.25	0.125	2	4.5
M30	18	140	0.125	0.25	0.25	0.125	0.5	0.25	0.125	2	4
M30	19	125	0.125	0.25	0.25	0.125	0.5	0.25	0.125	2	4.5
M30	20	100	0.125	0.25	0.25	0.125	0.5	0.25	0.125	2	4.5
M30	21	75	0.125	0.25	0.25	0.125	0.5	0.25	0.125	2	4.5
M30	22	50	0.125	0.25	0.25	0.125	0.5	0.25	2.125	2	4.5
M30	23	25	0.125	0.25	0.25	0.125	0.5	0.25	0.125	2	4.5
M30	24	10	0.125	0.25	0.25	0.125	0.5	0.25	0.125	2	4.5
M26a	1	1500	0.125		0.25	0.125	0.5	0.25	0.125	2	4.5
M26a	2	1300	0.125		0.25	0.125	0.5	0.25	0.125	2	4.5
M26a	3	1200	0.125		0.25	0.125	0.5	0.25	0.125	2	4.5
M26a	4	1100	0.125		0.25	0.125	0.5	0.25	0.125	2	4.5
M26a	5	1000	0.125		0.25	0.125	0.5	0.25	0.125	2	4.5
M26a	6	910	0.125		0.25	0.125	0.5	0.25	0.125	2	4.5
M26a	7	800	0.125		0.25	0.125	0.5	0.25	0.125	2	4.5
M26a	8	700	0.125		0.25	0.125	0.5	0.25	0.125	2	4.5
M26a	9	600	0.125		0.25	0.125	0.5	0.25	0.125	2	4.5
M26a	10	500	0.125		0.25	0.125	0.5	0.25	0.125	2	4.5
M26a	11	400	0.125		0.25	0.125	0.5	0.25	0.125	2	4.5
M26a	12	300	0.125		0.25	0.125	0.5	0.25	0.125	2	4.5
M26a	13	250	0.125		0.25	0.125	0.5	0.25	0.125	2	4.5
M26a	14	200	0.125		0.25	0.125	0.5	0.25	0.125	2	4.5
M26a	15	180	0.125		0.25	0.125	0.5	0.25	0.125	2	4.5
M26a	16	160	0.125		0.25	0.125	0.5	0.25	0.125	2	4.5
M26a	17	150	0.125	0.25	0.25	0.125	0.5	0.25	0.125	2	4.5
M26a	18	130	0.125	0.25	0.25	0.125	0.5	0.25	0.125	2	4.5
M26a	19	115	0.125	0.25	0.25	0.125	0.5	0.25	0.125	2	4.5
M26a	20	100	0.125	0.25	0.25	0.125	0.5	0.25	0.125	2	4.5

	M26a	21	75	0.125		0.25	0.25	0.125	0.5	0.25	0.125	2	4.5	
	M26a	22	50	0.125		0.25	0.25	0.125	0.5	0.25	2.125	2	4.5	
	M26a	23	25	0.125		0.25	0.25	0.125	0.5	0.25	0.125	2	4.5	
	M26a	24	10	0.125		0.25	0.25	0.125	0.5	0.25	0.125	2	4.5	
-	M22	1	400	0.125	0.25		0.25	0.125	0.5	0.25	0.125	2	4.5	
	M22	2	300	0.125	0.25		0.25	0.125	0.5	0.25	0.125	2	4.5	
	M22	3	270	0.125	0.25		0.25	0.125	0.5	0.25	0.125	2	4.5	
	M22	4	250	0.125	0.25		0.25	0.125	0.5	0.25	0.125	2	4.5	
	M22	5	230	0.125	0.25		0.25	0.125	0.5	0.25	0.125	2	4.5	
	M22	6	200	0.125	0.25		0.25	0.125	0.5	0.25	0.125	2	4.5	
	M22	7	180	0.125	0.25		0.25	0.125	0.5	0.25	0.125	2	4.5	
	M22	8	170	0.125	0.25		0.25	0.125	0.5	0.25	0.125	2	4.5	
	M22	9	160	0.125	0.25		0.25	0.125	0.5	0.25	0.125	2	4.5	
	M22	10	150	0.125	0.25	0.25	0.25	0.125	0.5	0.25	0.125	2	4.5	
	M22	11	140	0.125	0.25	0.25	0.25	0.125	0.5	0.25	0.125	2	4.5	
	M22	12	130	0.125	0.25	0.25	0.25	0.125	0.5	0.25	0.125	2	4.5	
	M22	13	120	0.125	0.25	0.25	0.25	0.125	0.5	0.25	0.125	2	4.5	
	M22	14	110	0.125	0.25	0.25	0.25	0.125	0.5	0.25	0.125	2	4.5	
	M22	15	100	0.125	0.25	0.25	0.25	0.125	0.5	0.25	0.125	2	4.5	
	M22	16	90	0.125	0.25	0.25	0.25	0.125	0.5	0.25	0.125	2	4.5	
	M22	17	80	0.125	0.25	0.25	0.25	0.125	0.5	0.25	0.125	2	4.5	
	M22	18	70	0.125	0.25	0.25	0.25	0.125	0.5	0.25	0.125	2	4.5	
	M22	19	60	0.125	0.25	0.25	0.25	0.125	0.5	0.25	0.125	2	4.5	
	M22	20	50	0.125	0.25	0.25	0.25	0.125	0.5	0.25	0.125	2	4.5	
	M22	21	40	0.125	0.25	0.25	0.25	0.125	0.5	0.25	0.125	2	4.5	
	M22	22	30	0.125	0.25	0.25	0.25	0.125	0.5	0.25	2.125	1	4.5	
	M22	23	20	0.125	0.25	0.25	0.25	0.125	0.5	0.25	0.125	2	4.5	
	M22	24	10	0.125	0.25	0.25	0.25	0.125	0.5	0.25	0.125	2	4.5	
	M22	1	5600	0.125	0.25		0.25	0.125	0.5	0.25	0.125	2	4.5	
	M22	2	5000	0.125	0.25		0.25	0.125	0.5	0.25	0.125	2	4.5	
	M22	3	4500	0.125	0.25		0.25	0.125	0.5	0.25	0.125	2	4.5	
	M22	4	4000	0.125	0.25		0.25	0.125	0.5	0.25	0.125	2	4.5	
	M22	5	3750											
	M22	6	3500	0.125	0.25		0.25	0.125	0.5	0.25	0.125	2	4.5	
	M22	7	3250	0.125	0.25		0.25	0.125	0.5	0.25	0.125	2	4	
	M22	8	3000	0.125	0.25		0.25	0.125	0.5	0.25	0.125	2	4.5	
	M22	9	2750	0.125	0.25		0.25	0.125	0.5	0.25	0.125	2	4.5	
	M22	10	2500	0.125	0.25		0.25	0.125	0.5	0.25	0.125	2	4.5	
	M22	11	2250	0.125	0.25		0.25	0.125	0.5	0.25	0.125	2	4.5	
	M22	12	2000	0.125	0.25		0.25	0.125	0.5	0.25	0.125	2	4.5	
	M22	13	1750	0.125	0.25		0.25	0.125	0.5	0.25	0.125	2	4.5	
	M22	14	1500	0.125	0.25		0.25	0.125	0.5	0.25	0.125	2	4.5	
	M22	15	1400	0.125	0.25		0.25	0.125	0.5	0.25	0.125	2	4.5	

Ν	/22 1	6 13	00 (	0.125	0.25		0.25	0.125	0.5	0.25	0.125	2	4.5
Ν	//22 1	7 12	00 0	0.125	0.25		0.25	0.125	0.5	0.25	0.125	2	4.5
Ν	//22 1	8 11	00 0	0.125	0.25		0.25	0.125	0.5	0.25	0.125	2	4.5
Ν	//22 1	9 10	00 0	0.125	0.25		0.25	0.125	0.5	0.25	0.125	2	4.5
Ν	/22 2	0 90	00 0	0.125	0.25		0.25	0.125	0.5	0.25	0.125	2	4.5
Ν	/22 2	1 80	)0 (	0.125	0.25		0.25	0.125	0.5	0.25	0.125	2	4.5
Ν	/22 2	2 70	00 0	0.125	0.25		0.25	0.125	0.5	0.25	2.125	1	4.5
Ν	/22 2	3 60	00 0	0.125	0.25		0.25	0.125	0.5	0.25	0.125	2	4.5
Ν	/22 2	4 50	00 0	0.125	0.25		0.25	0.125	0.5	0.25	0.125	2	4.5
N	/18	1 49	00 (	0.125			0.25	0.125	0.5	0.25	0.125	2	4.5
Ν	/18 2	2 45	00 (	0.125			0.25	0.125	0.5	0.25	0.125	2	4.5
Ν	/18	3 40	00 0	0.125			0.25	0.125	0.5	0.25	0.125	2	4.5
Ν	/18 4	4 35	00 (	0.125			0.25	0.125	0.5	0.25	0.125	2	4.5
Ν	/18 :	5 30	00 0	0.125			0.25	0.125	0.5	0.25	0.125	2	4.5
Ν	/18	5 25	00 0	0.125			0.25	0.125	0.5	0.25	0.125	2	4.5
Ν	/18	7 20	00 0	0.125			0.25	0.125	0.5	0.25	0.125	2	4.5
Ν	/18	3 15	00 0	0.125			0.25	0.125	0.5	0.25	0.125	2	4.5
Ν	/18	9 12	50 (	0.125			0.25	0.125	0.5	0.25	0.125	2	4.5
Ν	/18 1	0 10	00 0	0.125			0.25	0.125	0.5	0.25	0.125	2	4.5
Ν	/18 1	1 80	00 0	0.125			0.25	0.125	0.5	0.25	0.125	2	4.5
Ν	/18 1	2 60	00 0	0.125			0.25	0.125	0.5	0.25	0.125	2	4.5
Ν	/18 1	3 50	00 0	0.125			0.25	0.125	0.5	0.25	0.125	2	4.5
Ν	/18 1	4 40	00 0	0.125			0.25	0.125	0.5	0.25	0.125	2	4.5
Ν	/18 1	5 30	00 0	0.125			0.25	0.125	0.5	0.25	0.125	2	4.5
Ν	/18 1	6 20	00 0	0.125			0.25	0.125	0.5	0.25	0.125	2	4.5
Ν	/18 1	7 17	75 (	0.125			0.25	0.125	0.5	0.25	0.125	2	4.5
Ν	/18 1	8 15	55 (	0.125		0.25	0.25	0.125	0.5	0.25	0.125	2	4.5
Ν	/18 1	9 12	25 (	0.125		0.25	0.25	0.125	0.5	0.25	0.125	2	4.5
Ν	/18 2	0 10	)0 (	0.125		0.25	0.25	0.125	0.5	0.25	0.125	2	4.5
Ν	/18 2	1 7	5 (	0.125		0.25	0.25	0.125	0.5	0.25	0.125	2	4.5
Ν	/18 2	2 5	0 (	0.125		0.25	0.25	0.125	0.5	0.25	2.125	1	4.5
Ν	/18 2	3 2	5 (	0.125		0.25	0.25	0.125	0.5	0.25	0.125	2	4.5
N	И18 2	4 1	0 (	0.125		0.25	0.25	0.125	0.5	0.25	0.125	2	4.5
٢	<8a	1 40	00 (	0.125			0.25	0.125	0.5	0.25	0.125	2	4.5
٢	K8a 2	2 30	)0 (	0.125			0.25	0.125	0.5	0.25	0.125	2	4.5
ŀ	K8a	3 27	75 (	0.125			0.25	0.125	0.5	0.25	0.125	2	4.5
ŀ	<8a ·	4 25	50 (	0.125			0.25	0.125	0.5	0.25	0.125	2	4.5
ŀ	<8a :	5 23	30 (	0.125			0.25	0.125	0.5	0.25	0.125	2	4.5
ŀ	<8a (	5 20	00 0	0.125			0.25	0.125	0.5	0.25	0.125	2	4.5
ŀ	<8a ,	7 18	30 (	0.125			0.25	0.125	0.5	0.25	0.125	2	2.5
ŀ	<8a 3	8 17	70 (	0.125			0.25	0.125	0.5	0.25	0.125	2	4.5
٢	<8a	9 16	50 (	0.125			0.25	0.125	0.5	0.25	0.125	2	4.5
ŀ	K8a 1	0 15	50 (	0.125		0.25	0.25	0.125	0.5	0.25	0.125	2	4.5

K8a	11	140	0.125	0.25	0.25	0.125	0.5	0.25	0.125	2	4.5
K8a	12	130	0.125	0.25	0.25	0.125	0.5	0.25	0.125	2	4.5
K8a	13	120	0.125	0.25	0.25	0.125	0.5	0.25	0.125	2	4.5
K8a	14	110	0.125	0.25	0.25	0.125	0.5	0.25	0.125	2	4.5
K8a	15	100	0.125	0.25	0.25	0.125	0.5	0.25	0.125	2	4.5
K8a	16	90	0.125	0.25	0.25	0.125	0.5	0.25	0.125	2	4.5
K8a	17	80	0.125	0.25	0.25	0.125	0.5	0.25	0.125	2	4.5
K8a	18	70	0.125	0.25	0.25	0.125	0.5	0.25	0.125	2	4
K8a	19	60	0.125	0.25	0.25	0.125	0.5	0.25	0.125	2	4.5
K8a	20	50	0.125	0.25	0.25	0.125	0.5	0.25	0.125	2	4.5
K8a	21	40	0.125	0.25	0.25	0.125	0.5	0.25	0.125	2	4.5
K8a	22	30	0.125	0.25	0.25	0.125	0.5	0.25	2.125	1	4.5
K8a	23	20	0.125	0.25	0.25	0.125	0.5	0.25	0.125	2	4.5
K8a	24	10	0.125	0.25	0.25	0.125	0.5	0.25	0.125	2	4.5
K8a	1	5800	0.125		0.25	0.125	0.5	0.25	0.125	2	4.5
K8a	2	5500	0.125		0.25	0.125	0.5	0.25	0.125	2	4.5
K8a	3	5000	0.125		0.25	0.125	0.5	0.25	0.125	2	4.5
K8a	4	4500	0.125		0.25	0.125	0.5	0.25	0.125	2	4.5
K8a	5	4000	0.125		0.25	0.125	0.5	0.25	0.125	2	1.5
K8a	6	3500	0.125		0.25	0.125	0.5	0.25	0.125	2	4.5
K8a	7	3250	0.125		0.25	0.125	0.5	0.25	0.125	2	4.5
K8a	8	3000	0.125		0.25	0.125	0.5	0.25	0.125	2	4.5
K8a	9	2750	0.125		0.25	0.125	0.5	0.25	0.125	2	4.5
K8a	10	2500	0.125		0.25	0.125	0.5	0.25	0.125	2	4.5
K8a	11	2250	0.125		0.25	0.125	0.5	0.25	0.125	2	4.5
K8a	12	2000	0.125		0.25	0.125	0.5	0.25	0.125	2	4.5
K8a	13	1750	0.125		0.25	0.125	0.5	0.25	0.125	2	4.5
K8a	14	1500	0.125		0.25	0.125	0.5	0.25	0.125	2	4.5
K8a	15	1400	0.125		0.25	0.125	0.5	0.25	0.125	2	4.5
K8a	16	1300	0.125		0.25	0.125	0.5	0.25	0.125	2	4.5
K8a	17	1200	0.125		0.25	0.125	0.5	0.25	0.125	2	4.5
K8a	18	1100	0.125		0.25	0.125	0.5	0.25	0.125	2	4.5
K8a	19	1000	0.125		0.25	0.125	0.5	0.25	0.125	2	4.5
K8a	20	900	0.125		0.25	0.125	0.5	0.25	0.125	2	4.5
K8a	21	800	0.125		0.25	0.125	0.5	0.25	0.125	2	4.5
K8a	22	700	0.125		0.25	0.125	0.5	0.25	2.125	1	4.5
K8a	23	600	0.125		0.25	0.125	0.5	0.25	0.125	2	4.5
K8a	24	500	0.125		0.25	0.125	0.5	0.25	0.125	2	4.5
К9а	1	1000	0.125		0.25	0.125	0.5	0.25	0.125	2	2.5
К9а	2	800	0.125		0.25	0.125	0.5	0.25	0.125	2	2.5
К9а	3	700	0.125		0.25	0.125	0.5	0.25	0.125	2	2.5
K9a	4	600	0.125		0.25	0.125	0.5	0.25	0.125	2	2.5
K9a	5	500	0.125		0.25	0.125	0.5	0.25	0.125	2	2.5

K9a	6	400	0.125		0.25	0.125	0.5	0.25	0.125	2	2.5
K9a	7	300	0.125		0.25	0.125	0.5	0.25	0.125	2	2.5
K9a	8	250	0.125		0.25	0.125	0.5	0.25	0.125	2	2.5
K9a	9	200	0.125		0.25	0.125	0.5	0.25	0.125	2	2.5
K9a	10	180	0.125		0.25	0.125	0.5	0.25	0.125	2	2.5
K9a	11	170	0.125		0.25	0.125	0.5	0.25	0.125	2	2.5
K9a	12	160	0.125		0.25	0.125	0.5	0.25	0.125	2	2.5
K9a	13	150	0.125	0.25	0.25	0.125	0.5	0.25	0.125	2	2.5
K9a	14	140	0.125	0.25	0.25	0.125	0.5	0.25	0.125	2	2.5
K9a	15	130	0.125	0.25	0.25	0.125	0.5	0.25	0.125	2	2.5
K9a	16	120	0.125	0.25	0.25	0.125	0.5	0.25	0.125	2	2.5
K9a	17	110	0.125	0.25	0.25	0.125	0.5	0.25	0.125	2	2.5
K9a	18	100	0.125	0.25	0.25	0.125	0.5	0.25	0.125	2	2.5
K9a	19	90	0.125	0.25	0.25	0.125	0.5	0.25	0.125	2	2.5
K9a	20	80	0.125	0.25	0.25	0.125	0.5	0.25	0.125	2	2.5
K9a	21	70	0.125	0.25	0.25	0.125	0.5	0.25	0.125	2	2.5
K9a	22	60	0.125	0.25	0.25	0.125	0.5	0.25	2.125	1	2.5
K9a	23	30	0.125	0.25	0.25	0.125	0.5	0.25	0.125	2	2.5
K9a	24	5	0.125	0.25	0.25	0.125	0.5	0.25	0.125	2	2.5
K11a	1	1000	0.125		0.25	0.125	0.5	0.25	0.125	2	2.5
K11a	2	800	0.125		0.25	0.125	0.5	0.25	0.125	2	2.5
K11a	3	650	0.125		0.25	0.125	0.5	0.25	0.125	2	2.5
K11a	4	500	0.125		0.25	0.125	0.5	0.25	0.125	2	2.5
K11a	5	350	0.125		0.25	0.125	0.5	0.25	0.125	2	2.5
K11a	6	200	0.125		0.25	0.125	0.5	0.25	0.125	2	2.5
K11a	7	175	0.125		0.25	0.125	0.5	0.25	0.125	2	2.5
K11a	18	140	0.125	0.25	0.25	0.125	0.5	0.25	0.125	2	2.5
K11a	19	125	0.125	0.25	0.25	0.125	0.5	0.25	0.125	2	2.5
K11a	20	100	0.125	0.25	0.25	0.125	0.5	0.25	0.125	2	2.5
K11a	21	75	0.125	0.25	0.25	0.125	0.5	0.25	0.125	2	2.5
K11a	22	50	0.125	0.25	0.25	0.125	0.5	0.25	2.125	1	2.5
K11a	23	25	0.125	0.25	0.25	0.125	0.5	0.25	0.125	2	2.5
K11a	24	10	0.125	0.25	0.25	0.125	0.5	0.25	0.125	2	2.5
K12a	1	4413	0.125		0.25	0.125	0.5	0.25	0.125	2	2.5
K12a	2	4413	0.125		0.25	0.125	0.5	0.25	0.125	2	2.5
K12a	3	4000	0.125		0.25	0.125	0.5	0.25	0.125	2	2.5
K12a	4	3500	0.125		0.25	0.125	0.5	0.25	0.125	2	2.5
K12a	5	3000	0.125		0.25	0.125	0.5	0.25	0.125	2	2.5
K12a	6	2500	0.125		0.25	0.125	0.5	0.25	0.125	2	2.5
K12a	7	2000	0.125		0.25	0.125	0.5	0.25	0.125	2	2.5
K12a	8	1500	0.125		0.25	0.125	0.5	0.25	0.125	2	2.5
K12a	9	1000	0.125		0.25	0.125	0.5	0.25	0.125	2	2.5
K12a	10	800	0.125		0.25	0.125	0.5	0.25	0.125	2	2.5

K12a	11	600	0.125		0.25	0.125	0.5	0.25	0.125	2	2.5
K12a	12	500	0.125		0.25	0.125	0.5	0.25	0.125	2	2.5
K12a	13	400	0.125		0.25	0.125	0.5	0.25	0.125	2	2.5
K12a	14	300	0.125		0.25	0.125	0.5	0.25	0.125	2	2.5
K12a	15	200	0.125		0.25	0.125	0.5	0.25	0.125	2	2.5
K12a	16	180	0.125		0.25	0.125	0.5	0.25	0.125	2	2.5
K12a	17	160	0.125		0.25	0.125	0.5	0.25	0.125	2	2.5
K12a	18	150	0.125	0.25	0.25	0.125	0.5	0.25	0.125	2	2.5
K12a	19	130	0.125	0.25	0.25	0.125	0.5	0.25	0.125	2	2.5
K12a	20	100	0.125	0.25	0.25	0.125	0.5	0.25	0.125	2	2.5
K12a	21	75	0.125	0.25	0.25	0.125	0.5	0.25	0.125	2	2.5
K12a	22	50	0.125	0.25	0.25	0.125	0.5	0.25	2.125	1	2.5
K12a	23	25	0.125	0.25	0.25	0.125	0.5	0.25	0.125	2	2.5
K12a	24	10	0.125	0.25	0.25	0.125	0.5	0.25	0.125	2	2.5
K13	1	1000									
K13	2	800			0.25	0.125	0.5	0.25	0.125	2	1.5
K13	3	700			0.25	0.125	0.5	0.25	0.125	2	1.5
K13	4	600			0.25	0.125	0.5	0.25	0.125	2	1.5
K13	5	500									
K13	6	400			0.25	0.125	0.5	0.25	0.125	2	1.5
K13	7	300			0.25	0.125	0.5	0.25	0.125	2	1.5
K13	8	250			0.25	0.125	0.5	0.25	0.125	2	1.5
K13	9	200			0.25	0.125	0.5	0.25	0.125	2	1.5
K13	10	180			0.25	0.125	0.5	0.25	0.125	2	1.5
K13	11	170			0.25	0.125	0.5	0.25	0.125	2	1.5
K13	12	160			0.25	0.125	0.5	0.25	0.125	2	1.5
K13	13	150			0.25	0.125	0.5	0.25	0.125	2	1.5
K13	14	140			0.25	0.125	0.5	0.25	0.125	2	1.5
K13	15	130			0.25	0.125	0.5	0.25	0.125	2	1.5
K13	16	120			0.25	0.125	0.5	0.25	0.125	2	1.5
K13	17	110									
K13	18	100									
K13	19	90			0.25	0.125	0.5	0.25	0.125	2	1.5
K13	20	80			0.25	0.125	0.5	0.25	0.125	2	1.5
K13	21	70			0.25	0.125	0.5	0.25	0.125	2	1.5
K13	22	60			0.25	0.125	0.5	0.25	2.125	1	1.5
K13	23	30			0.25	0.125	0.5	0.25	0.125	2	1.5
K13	24	5			0.25	0.125	0.5	0.25	0.125	2	1.5

(note : The blank means no sampling, the"-----"means no collecting water )

#### References

- Brown, M. T., and K. W. Bruland. 2008. An improved flow-injection analysis method for the determination of dissolved aluminum in seawater. Limnology and Oceanography: Methods, 6(1): 87-95.
- Aguilar-Islas A. M., and K. W. Bruland. 2006. Dissolved manganese and silicic acid in the Columbia River plume: a major source to the California current and coastal waters off Washington and Oregon. Marine chemistry, 101(3-4): 233-247.
- Conway T. M., A. D. Rosenberg, J. F. Adkins, et al. 2013. A new method for precise determination of iron, zinc and cadmium stable isotope ratios in seawater by double-spike mass spectrometry. Analytica Chimica Acta, 793: 44-52.

#### 4.3.6. REE (Prof. Zhang)

The REE is widely used as water mass tracer in the different ocean basins. In our study, we will trace sources of macro- and micro-nutrients to the North Pacific Subtropical Gyre from terrestrial input and water mass transport, using REE. In this cruise, the seawater samples of REE were taken by Clean Niskin bottles. 500 mL of seawater were filtered in-line from Niskin-X bottles in the clean van through AcroPak 1000 cartridges (pore size of  $0.8/0.45 \ \mu m$ ) into pre-cleaned LDPE bottles.

REE Seawater samples were taken from Clean CTD at Stations K2b, KPR, WPS, MR04, MR05, M35, M30, M26a, M22, M18, K8a, K9a, K11a, K12a and K13 from surface to bottom. The unfiltered clean seawater samples of REEs were sampled at the K12a station. The samples were acidified to a pH<2 with 1ml ultra-clean 6 N HCl (optima grade). The concentration of REEs will be treated using NOBIAS Resin, and then measured by ICP-MS.

#### 4.3.7. Al/Mn

#### **Objectives:**

- 1. To determine the concentrations of the dissolved aluminum (dAl) and dissolved manganese (dMn) in North Pacific Subtropical Gyre (NPSG).
- 2. To improve the understanding of biogeochemical cycles of dAl and dMn in NPSG.
- 3. To trace the sources of micro-nutrients from terrestrial input, atmospheric deposition and hydrothermal vent etc. to NPSG.

#### **Analytical Methods:**

The concentrations of dAl and dMn will be analyzed by the flow injection analysis methods with on-line preconcentration following the protocol described in Brown and Bruland (2008) and Aguilar-Islas and Bruland (2006). The detection limits for dAl and dMn are 0.18 nmol  $L^{-1}$  and 0.2 nmol  $L^{-1}$ , respectively. The analytical precision is better than 4% for dAl, 5% for dMn.

#### **Sampling information:**

Samples for trace elements were collected by 12-liter Niskin-X bottles mounted onto a clean rosette sampling assembly, equipped with a CTD recorder (Sea-Bird SBE911Plus).

Unfiltered and filtered seawater samples were subsampled for each depth. More detailed sampling information is given in Table 8. To obtain filtered samples, seawater was filtered in-line from Niskin-X bottles in the clean van through AcroPak 1000 cartridges (pore size of  $0.8/0.45 \,\mu\text{m}$ ) into pre-cleaned LDPE bottles. The samples were acidified to a pH of ~1.8 with ultra-clean HCl (Merck) in a 100-class clean bench in the laboratory and stored for more than one month before analysis.

Parameter	Sample station	Sample	Sample
1 drumeter	Sumple Station	Volume	counts
Filtered-dAl/ dMn (Clean CTD)	K2b, KPR, WPS, MR04, MR05, M35, M30, M26a, M22, M18, K8a, K9a, K11a, K12a, K13	250ml	426
Unfiltered- dAl/ dMn (Clean CTD)	K2b, KPR, WPS, M22, K12a	250ml	168

Table 8 Sampling information for dAl and dMn

#### References

- Brown, M. T., and K. W. Bruland. 2008. An improved flow-injection analysis method for the determination of dissolved aluminum in seawater. Limnology and Oceanography: Methods, 6(1): 87-95.
- Aguilar-Islas A. M., and K. W. Bruland. 2006. Dissolved manganese and silicic acid in the Columbia River plume: a major source to the California current and coastal waters off Washington and Oregon. Marine chemistry, 101(3-4): 233-247.

#### 4.4 Towed fish (Trace Metal Clean Underway) (Profs. Cai and Zhang)

#### 4.4.1. Objectives

The aim of the towed fish sampling in this cruise was to obtain the high spatial resolution biogeochemical information along the cruise track in the wNP. The main objective of the GEOTRACES program was to describe the high-resolution trace element distribution in the global ocean. While the trace metal clean rosettes sampling allows us to obtain the high resolution vertical distribution of TEIs, the towed fish sampling allows us to observe a high resolution surface distribution for the trace substances in the ocean. Based on the information from the fish sampling, we can depict the distribution patterns of a lot more detailed biogeochemical parameters along the cruise track, and are able to link the detailed physical, biological, and chemical processes at surface. The fish also provided an opportunity to collect trace metal clean surface seawater for the incubation experiments.

#### 4.4.2. Parameters

1) Underway samples were collected at a high spatial resolution ( $\sim$ 50km) by a towed fish while the ship was steaming. Trace metals (Fe, Cu, Zn, Cd, Ni, Pb), trace mental isotopes ( $\delta$ 56Fe,

#### δ114Cd)

2) Particulate organic carbon and lipid biomarkers

#### 4.4.3. Analytical methods

The concentration of dissolved Fe, Cu, Zn, Cd, Ni, Pb will be pre-concentrated and analyzed by inductivity coupled plasma mass spectrometry (ICP-MS, Thermo ICAP-Q) using isotopic dilution or standard addition method. The analytical methods for trace metal isotopes are described at sections 1.3. POC and lipid biomarker analysis were carried out according to the procedures described by Bi et al. (2018) and Wang et al. (2019). A piece of freeze-dried sample filter was used for POC analysis. Carbonate was removed by adding 6 mol L<sup>-1</sup> HCl, then drying at 55 °C before conducting elemental analysis with a Thermo Flash 2000 Elemental Analyzer. The precision for POC analysis was  $\pm 0.02\%$  (wt.%, n = 6) in our laboratory, determined by replicate analysis of atropine (Thermo Fisher Scientific, Netherlands) and a low organic content soil (Elemental Microanalysis Ltd., UK).

Suspended particle samples for lipid biomarker analysis were freeze-dried, then ultrasonically extracted with a 3:1 (v/v) mixture of dichloromethane and methanol after adding internal standards i.e., C19 n-alkanol. Total lipid extracts were concentrated under a stream of N2 gas and saponified with 6% KOH in methanol overnight. The neutral component was then extracted with n-hexane and concentrated under N2. The extracts were subsequently separated into fractions using silica gel chromatography. The neutral lipid fraction containing brassicasterol, dinosterol, and C37 alkenones was eluted with 22 mL dichloromethane/methanol (95:5, vol/vol), dried under a gentle N2 stream, and derivatized using N, O-bis(trimethylsily)-trifluoroacetamide at 70°C for 1 h before instrumental measurements. Biomarker quantification was analyzed on an Agilent 7890N Gas-Chromatograph with an HP-1 capillary column (50 m × 0.32  $\mu$ m × 0.17  $\mu$ m) and a flame ionization detector. The oven temperature program was as follows: 80 °C for 1 min initially, increasing to 200 °C at 25 °C min<sup>-1</sup>, then increasing to 250 °C at 4 °C min<sup>-1</sup>, then increasing to 300 °C at 1.7 °C min<sup>-1</sup> and holding for 15 min, and finally increasing to 315 °C at 5 °C min<sup>-1</sup> and holding for 5 min. Lipid biomarkers were quantified by compare GC-FID peak areas to those of the internal standard C19 n-alkanol.

#### References

- Bi, R., Chen, X., Zhang, J., Ishizaka, J., Zhuang, Y., Jin, H., et al. (2018). Water mass control on phytoplankton spatiotemporal variations in the northeastern East China Sea and the western Tsushima Strait revealed by lipid biomarkers. Journal of Geophysical Research: Biogeosciences, 123, 1318–1332. https://doi.org/10.1002/ 2017JG004340.
- Wang, Y., D.-W. Li, J. P. Sachs, J. Hu, Y. Cao, L. Li, H. Zhang, and M. Zhao (2019), Vertical distribution of isoprenoid GDGTs in suspended particles from the East China Sea shelf and implications for sedimentary TEX<sup>H</sup><sub>86</sub> records, Organic Geochemistry, 136, 103895, doi:10.1016/j.orggeochem.2019.07.004.

#### 4.4.4. Sampling information

A towed fish was deployed to obtain high resolution seawater samples. The configuration of the fish system was upgraded from the combination of the previous designs, e.g. Bruland et al. (2005), Vink et al. (2000) and Zhang et al. (2019). A 6-m long boom kept the fish distanced from the ship when towed at sea. Low density polyethylene tubing was used to collected surface samples from the sea surface directly into a trace mental clean van on the vessel. Our fish system allowed collection of trace mental clean seawater at a depths between 0.5-1.5m while the ship speed was at 10-13 knots during the whole cruise. The samples were either collected unfiltered, or filtered by a  $0.8/0.2 \mu$ m capsule filter (Millipore). A total of 162 surface towed fish samples were obtained by the towed fish system. The fish system was first deployed at 18:00, 7/4/2020 in the oligotrophic Luzon Strait, and recovered at 23:00, 8/20/2020 in the coastal ocean of Taiwan Strait.

		are to not the mon sumpres
Sample ID	Date (Beijing time)	Time (Beijing time)
kk2003-CF-0001	2020/7/4	18:00
kk2003-CF-0002	2020/7/4	21:00
kk2003-CF-0003	2020/7/5	0:00
kk2003-CF-0004	2020/7/5	3:00
kk2003-CF-0005	2020/7/5	6:00
kk2003-CF-0006	2020/7/5	9:00
kk2003-CF-0007	2020/7/5	12:00
kk2003-CF-0008	2020/7/5	15:00
kk2003-CF-0009	2020/7/5	18:00
kk2003-CF-0010	2020/7/5	21:00
kk2003-CF-0011	2020/7/9	12:00
kk2003-CF-0012	2020/7/9	15:00
kk2003-CF-0013	2020/7/9	18:00
kk2003-CF-0014	2020/7/9	21:00
kk2003-CF-0015	2020/7/10	0:00
kk2003-CF-0016	2020/7/10	3:00
kk2003-CF-0017	2020/7/10	6:00
kk2003-CF-0018	2020/7/10	9:00
kk2003-CF-0019	2020/7/10	12:00
kk2003-CF-0020	2020/7/10	15:00
kk2003-CF-0021	2020/7/10	18:00
kk2003-CF-0022	2020/7/10	21:00
kk2003-CF-0023	2020/7/11	0:00
kk2003-CF-0024	2020/7/11	3:00
kk2003-CF-0025	2020/7/11	6:00
kk2003-CF-0026	2020/7/11	9:00
kk2003-CF-0027	2020/7/11	12:00

Table 9 The information of 162 surface towed fish samples

kk2003-CF-0028	2020/7/11	15:00
kk2003-CF-0029	2020/7/11	18:00
kk2003-CF-0030	2020/7/11	21:00
kk2003-CF-0031	2020/7/12	9:00
kk2003-CF-0032	2020/7/12	12:30
kk2003-CF-0033	2020/7/12	15:35
kk2003-CF-0034	2020/7/12	18:00
kk2003-CF-0035	2020/7/12	21:00
kk2003-CF-0036	2020/7/13	0:00
kk2003-CF-0037	2020/7/13	9:00
kk2003-CF-0038	2020/7/13	12:00
kk2003-CF-0039	2020/7/13	15:00
kk2003-CF-0040	2020/7/13	18:00
kk2003-CF-0041	2020/7/13	21:00
kk2003-CF-0042	2020/7/14	0:00
kk2003-CF-0043	2020/7/20	6:00
kk2003-CF-0044	2020/7/20	9:00
kk2003-CF-0045	2020/7/20	12:00
kk2003-CF-0046	2020/7/20	15:00
kk2003-CF-0047	2020/7/21	21:00
kk2003-CF-0048	2020/7/22	0:00
kk2003-CF-0049	2020/7/22	3:00
kk2003-CF-0050	2020/7/22	6:00
kk2003-CF-0051	2020/7/22	9:00
kk2003-CF-0052	2020/7/22	12:00
kk2003-CF-0053	2020/7/22	15:00
kk2003-CF-0054	2020/7/22	18:00
kk2003-CF-0055	2020/7/22	21:00
kk2003-CF-0056	2020/7/23	0:00
kk2003-CF-0057	2020/7/23	3:00
kk2003-CF-0058	2020/7/23	6:00
kk2003-CF-0059	2020/7/23	9:00
kk2003-CF-0060	2020/7/23	12:00
kk2003-CF-0061	2020/7/23	15:00
kk2003-CF-0062	2020/7/23	18:00
kk2003-CF-0063	2020/7/23	21:00
kk2003-CF-0064	2020/7/26	15:00
kk2003-CF-0065	2020/7/26	18:00
kk2003-CF-0066	2020/7/26	21:30
kk2003-CF-0067	2020/7/27	0:00
kk2003-CF-0068	2020/7/27	3:00
kk2003-CF-0069	2020/7/27	12:00

kk2003-CF-0070	2020/7/27	15:00
kk2003-CF-0071	2020/7/27	18:00
kk2003-CF-0072	2020/7/28	9:00
kk2003-CF-0073	2020/7/28	12:00
kk2003-CF-0074	2020/7/28	15:00
kk2003-CF-0075	2020/7/28	18:00
kk2003-CF-0076	2020/7/28	21:00
kk2003-CF-0077	2020/7/29	0:00
kk2003-CF-0078	2020/7/29	3:00
kk2003-CF-0079	2020/7/29	6:00
kk2003-CF-0080	2020/7/29	9:00
kk2003-CF-0081	2020/7/29	12:00
kk2003-CF-0082	2020/7/29	16:40
kk2003-CF-0083	2020/7/29	18:00
kk2003-CF-0084	2020/7/31	9:00
kk2003-CF-0085	2020/7/31	12:00
kk2003-CF-0086	2020/7/31	15:00
kk2003-CF-0087	2020/7/31	18:00
kk2003-CF-0088	2020/7/31	21:00
kk2003-CF-0089	2020/8/1	0:00
kk2003-CF-0090	2020/8/1	3:00
kk2003-CF-0091	2020/8/1	6:00
kk2003-CF-0092	2020/8/1	9:00
kk2003-CF-0093	2020/8/1	12:00
kk2003-CF-0094	2020/8/4	0:00
kk2003-CF-0095	2020/8/4	3:00
kk2003-CF-0096	2020/8/4	6:00
kk2003-CF-0097	2020/8/4	9:00
kk2003-CF-0098	2020/8/4	12:00
kk2003-CF-0099	2020/8/4	21:00
kk2003-CF-0100	2020/8/5	0:00
kk2003-CF-0101	2020/8/5	3:00
kk2003-CF-0102	2020/8/6	15:00
kk2003-CF-0103	2020/8/6	18:00
kk2003-CF-0104	2020/8/6	21:00
kk2003-CF-0105	2020/8/7	0:00
kk2003-CF-0106	2020/8/7	6:00
kk2003-CF-0107	2020/8/7	9:00
kk2003-CF-0108	2020/8/7	12:00
kk2003-CF-0109	2020/8/7	15:00
kk2003-CF-0110	2020/8/10	9:00
kk2003-CF-0111	2020/8/10	12:00

kk2003-CF-0112	2020/8/10	15:00
kk2003-CF-0113	2020/8/10	18:00
kk2003-CF-0114	2020/8/10	21:00
kk2003-CF-0115	2020/8/11	0:00
kk2003-CF-0116	2020/8/11	3:00
kk2003-CF-0117	2020/8/11	6:00
kk2003-CF-0118	2020/8/11	9:30
kk2003-CF-0119	2020/8/14	6:00
kk2003-CF-0120	2020/8/14	9:00
kk2003-CF-0121	2020/8/14	12:00
kk2003-CF-0122	2020/8/14	15:30
kk2003-CF-0123	2020/8/14	18:00
kk2003-CF-0124	2020/8/14	22:00
kk2003-CF-0125	2020/8/15	0:00
kk2003-CF-0126	2020/8/15	3:00
kk2003-CF-0127	2020/8/15	6:00
kk2003-CF-0128	2020/8/15	9:00
kk2003-CF-0129	2020/8/16	0:00
kk2003-CF-0130	2020/8/16	3:00
kk2003-CF-0131	2020/8/16	6:00
kk2003-CF-0132	2020/8/16	9:00
kk2003-CF-0133	2020/8/16	12:00
kk2003-CF-0134	2020/8/16	15:00
kk2003-CF-0135	2020/8/16	18:00
kk2003-CF-0136	2020/8/16	21:00
kk2003-CF-0137	2020/8/17	0:00
kk2003-CF-0138	2020/8/18	0:00
kk2003-CF-0139	2020/8/18	3:00
kk2003-CF-0140	2020/8/18	6:00
kk2003-CF-0141	2020/8/18	9:00
kk2003-CF-0142	2020/8/18	12:00
kk2003-CF-0143	2020/8/18	15:00
kk2003-CF-0144	2020/8/18	18:00
kk2003-CF-0145	2020/8/18	21:00
kk2003-CF-0146	2020/8/19	0:00
kk2003-CF-0147	2020/8/19	3:00
kk2003-CF-0148	2020/8/19	6:00
kk2003-CF-0149	2020/8/19	9:00
kk2003-CF-0150	2020/8/19	12:00
kk2003-CF-0151	2020/8/19	15:00
kk2003-CF-0152	2020/8/19	18:00
kk2003-CF-0153	2020/8/19	21:00

kk2003-CF-0154	2020/8/20	0:00
kk2003-CF-0155	2020/8/20	3:00
kk2003-CF-0156	2020/8/20	6:00
kk2003-CF-0157	2020/8/20	9:30
kk2003-CF-0158	2020/8/20	12:00
kk2003-CF-0159	2020/8/20	15:00
kk2003-CF-0160	2020/8/20	18:00
kk2003-CF-0161	2020/8/20	21:00
kk2003-CF-0162	2020/8/20	23:00

We used FIA method to measure the dFe concentrations onboard. Suspended particle samples for POC and lipid biomarker analyses were collected from surface seawater at 18 stations, obtained by filtration (water volume: 260–300 L) on pre-combusted glass microfiber filters (Whatman GF/F filters, 0.7  $\mu$ m pore size) and stored at –20 °C until laboratory analysis.

		1 0		<u>.</u>	
Station	Sampling date	Latitude (°N)	Longitude (°E)	Sample depth (m)	Water volume (L)
K2b	20200706	20.35	126.27	3	300
KPR	20200712	16.52	135.02	3	260
WPS	20200715	20	140	3	280
MR04	20200719	20	150	3	300
MR05	20200721	25	150	3	300
M35	20200724	33	155	3	300
M32	20200727	30.5	155	3	300
M30	20200728	28.48	154.84	3	300
M26a	20200731	24.02	157.96	3	260
M22	20200802	20.15	154.72	3	300
M18	20200805	16	154.83	3	300
K8a	20200808	12.53	154.74	3	300
K9a	20200811	13	150	1	300
K11a	20200813	14	140	1	300
K12a	20200815	12.78	134.98	1	300
K13	20200817	10.97	129.95	1	300
А	20200819	16.98-17.11	127.44-127.29	3	270
В	20200819	19.36	126.54	1	260

Table 10 Sampling information for POC and lipid biomarkers

#### **References:**

- Bruland, K. W., E. L. Rue, G. J. Smith, and G. R. DiTullio. 2005. Iron, macronutrients and diatom blooms in the Peru upwelling regime: brown and blue waters of Peru. Mar. Chem., 93: 81-103.
- Vink, S., E. Boyle, C. Measures, and J. Yuan. 2000. Automated high resolution determination of the trace elements iron and aluminium in the surface ocean using a towed Fish coupled to flow injection analysis. Deep-Sea Res.I, 47: 1141-1156.
- Zhang, R., X. Zhu, C. Yang, L. Ye, G. Zhang, J. Ren, Y. Wu, S. Liu, J. Zhang, and M. Zhou. 2019. Distribution of dissolved iron in the Pearl River (Zhujiang) Estuary and the northern continental slope of the South China Sea.

## 4.5. Chemical Oceanography On Normal CTD casts (Profs. Dai, Li , Guo, Cai, Cao, and Zhang)

#### 4.5.1. Nutrients

Nutrients samples were collected from 12-liter Niskin bottles mounted onto a Rosette sampling assembly, equipped with a CTD recorder (Sea-Bird SBE911 plus). Samples for nitrate  $(NO_3^{-})$ , nitrite  $(NO_2^{-})$ , soluble reactive phosphate (SRP), and silicate  $(Si(OH)_4)$  were collected using 125 mL high-density polyethylene bottles at all stations with full depth profiles, and for  $NO_3^-$ ,  $NO_2^-$  and SRP at nanomolar level were usually collected at all stations within upper 170 m. Samples for nutrients (at micro-molar level ) were stored at 4 °C and for nutrients at nanomolar level, frozen at -20 °C, until analysis. Nutrients from discrete samples were analyzed onboard with a Four-channel Continuous Flow Technicon AA3 Auto-Analyzer (Bran-Lube, GmbH) according to classic colorimetric methods (Becker et al., 2019). Nitrite reacts with sulfanilamide under acidic conditions resulting in а diazo compound form, which is coupled with N-1-naphthyl-ethylenediamine hydrochloride to form a reddish-purple azo dye. Nitrate is reduced to nitrite form under alkaline conditions using a copperized cadmium reductor coil. SRP and Si(OH)<sub>4</sub> were both measured using spectrophotometric methods with ammonium molybdate (Knap et al., 1996). The reliable quantitation limit (RQL) for NO<sub>3</sub><sup>-</sup>, NO<sub>2</sub><sup>-</sup>, SRP and Si(OH)<sub>4</sub> are 0.04 µmol L<sup>-1</sup>, 0.04 µmol L<sup>-1</sup>, 0.08 µmol L<sup>-1</sup> and 0.16 µmol L<sup>-1</sup>, respectively. The analytical precision is  $\pm 0.218\%$  for NO<sub>3</sub><sup>-</sup> at 10.4µmol/L,  $\pm 0.38\%$  for NO<sub>2</sub><sup>-</sup> at 0.4µmol/L,  $\pm 0.253\%$  for SRP at  $0.8\mu$ mol/L and  $\pm 0.022\%$  for Si(OH)<sub>4</sub> at 40 nmol/L. The measurement of NO<sub>3</sub><sup>-</sup> at nanomolar level was conducted on a continuous flow analysis system combined with a liquid waveguide capillary flow cell. The reliable quantitation limit (RQL) of NO3<sup>-</sup> at nanomolar level is 7.46 nmol L<sup>-1</sup> and the precision is better than  $\pm 10\%$ . SRP at nanomolar level was determined using a flow injection system and enrichment by HLB cartridge method. The reliable quantitation limit is 4.2 nmol  $L^{-1}$ and the precision is better than  $\pm 10\%$  (Ma et al., 2008).

#### References

- Becker, S., Aoyama, M., Woodward, E. M. S., Bakker, K., Coverly, S., Mahaffey, C., & Tanhua, T. (2020). GO-SHIP repeat hydrography nutrient manual: the precise and accurate determination of dissolved inorganic nutrients in seawater, using continuous flow analysis methods. *Frontiers in Marine Science*, 7, 908.
- Knap, A., Michaels, A., Close, A., Ducklow, H., & Dickson, A. (1996). Protocols for the joint global ocean flux study (JGOFS) core measurements. In. p. 43-90. JGOFS Report No. 19, vi + 170 pp. Reprint of the IOC Manuals and Guides No. 29, UNESCO 1994.
- Ma, J., Yuan, D., & Liang, Y. (2008). Sequential injection analysis of nanomolar soluble reactive phosphorus in seawater with HLB solid phase extraction. *Marine Chemistry*, 111(3-4), 151-159.

# 4.5.2. Dissolved Organic Carbon (DOC), Dissolved Organic Phosphorus and nitrogen (DOP & DON)

#### 4.5.2.1. Dissolved Organic Carbon

The collection and measurement of samples for DOC follow the JGOFS protocol (Benner and Strom, 1993; Sharp and Peltzer, 1993). Duplicate samples for DOC were collected from 12-liter Niskin bottles mounted onto a Rosette sampling assembly, equipped with a CTD recorder (Sea-Bird SBE911 plus), at all stations with full depth profiles. For the depths within the upper 200 m, filtered samples were also collected using on-line filter holder packaged with 450 °C baked GF/F membrane to eliminate the contribution from particulate organic carbon. Samples for DOC analysis were collected into 40 mL borosilicate glass bottles and stored at -20 °C until analysis. DOC will be measured using a total organic carbon analyzer (Model TOC-V<sub>CPH</sub>, Shimazu Co., Ltd.) with the method of high temperature combustion (HTC) followed by quantitative measurements of the CO<sub>2</sub> produced by non-dispersive infra-red (NDIR) analysis. The precision is  $\pm 1 \ \mu mol \ L^{-1}$ . DOC will be calibrated against certified reference material provided by Dr. Hansell, D.A. at the University of Miami.

#### References

Benner, R., & Strom, M. (1993). A critical evaluation of the analytical blank associated with doc measurements by high-temperature catalytic oxidation. *Marine Chemistry*, 41(1-3), 153-160.

Sharp, J. H., Peltzer, E. T., Alperin, M. J., Cauwet, G., & Carlson, C. A. (1993). Procedures subgroup report. *Marine Chemistry*, 41(s 1 - 3), 37-49.

#### 4.5.2.2. Dissolved Organic Phosphorus and nitrogen (DOP & DON)

Duplicate samples for DOP were collected with 12-liter Niskin bottles mounted onto a Rosette sampling assembly, equipped with a CTD recorder (Sea-Bird SBE911 plus) at all stations with full depth profiles. These samples are collected into 125 mL acid-cleaned (10% HCl) HDPE bottles and stored at -20°C until analysis. Total dissolved phosphorus (TDP) and Total dissolved nitrogen (TDN) will be determined by converting organic phosphorus and nitrogen compounds into orthophosphate and nitrate under high energy UV radiation, followed by nutrients measurement method above with UV-Vis spectrophotometry (Foreman et al., 2019). DOP is then calculated by subtracting dissolved inorganic phosphorus from TDP; and DON is calculated by subtracting dissolved inorganic nitrogen from TDN.

#### References

Foreman, R. K., Björkman, Karin M., Carlson, C. A., Opalk, K., & Karl, D. M. (2019). Improved ultraviolet photo-oxidation system yields estimates for deep-sea dissolved organic nitrogen and phosphorus. *Limnology and oceanography, methods*.

## 4.5.2.2. DOC and absorption spectroscopy and fluorescence spectroscopy for DOM (Prof. Zhang)

For DOM samples, seawater samples were filtered immediately through Whatman GF/F glass fiber filters (47 mm diameter) combusted at 450 °C, and then refiltered through polyethersulfone (PES; 25 mm diameter) syringe (0.22 µm porosity) filters. All of the DOM samples were stored at 4 °C in pre-cleaned and pre-combusted amber glass vials for subsequent laboratory analyses, such as acid washing and combustion at 450 °C for 4 h.

DOC was measured by high-temperature combustion with a Multi N/C 3100 TOC-TN analyzer (Analytik Jena, Germany). Samples were acidified with 3 M HCl and purged with oxygen to remove the inorganic carbon prior to analysis. Three 50  $\mu$ l aliquots were injected into a combustion tube filled with a platinum catalyst by using an auto sampler. The combusted products (carbon dioxide) were detected with a gas analyzer. The DOC concentration was determined using the calibration curve of the standard solution of potassium hydrogen phthalate. Absorption spectra were recorded from 250 nm to 700 nm by using UV–visible spectrophotometer (UV-2550, Shimadzu) equipped with a 10-cm quartz cell. Milli-Q water was used as blank.

EEMs were recorded using a Hitachi F-4500 fluorescence spectrophotometer (Hitachi) equipped with a 1cm quartz cell. Instrumental response was corrected according to the procedure recommended by the instruction manual of Hitachi F-4500. Seawater samples were taken from hydro CTD at Stations K2b, WPS, MR04, M35, M30, M22, M18, K8a and K13, from surface to bottom. The detailed information is presented in table below.

Station	Latitude	Longitude	Cast	Bottle	Depth
(°N)	(°N)	(°E)	Number	Number	(m)
K2b	20.59	126.22	19	1	5000
K2b	20.59	126.22	19	3	3000
K2b	20.59	126.22	19	7	1000
K2b	20.59	126.22	19	9	800
K2b	20.59	126.22	19	11	640
K2b	20.59	126.22	19	13	500
K2b	20.59	126.22	19	15	350
K2b	20.59	126.22	19	17	170
K2b	20.59	126.22	19	19	100
K2b	20.59	126.22	19	21	50
K2b	20.59	126.22	19	23	10
WPS	19.98	140.00	13	2	4600
WPS	19.98	140.00	13	4	3000
WPS	19.98	140.00	13	6	2000
WPS	19.98	140.00	13	8	1000
WPS	19.98	140.00	13	10	800
WPS	19.98	140.00	13	12	600

 Table 11 The detailed sampling information of Mercury species and DOM characteristics for

 hydro CTD

WPS	19.98	140.00	13	14	500
WPS	19.98	140.00	13	16	300
WPS	19.98	140.00	13	18	200
WPS	19.98	140.00	13	20	150
WPS	19.98	140.00	13	22	70
WPS	19.98	140.00	13	24	25
MR04	20.02	149.96	2	1	4600
MR04	20.02	149.96	2	5	2000
MR04	20.02	149.96	2	12	1000
MR04	20.02	149.96	2	16	800
MR04	20.02	149.96	2	20	600
MR04	20.02	149.96	2	23	500
MR04	20.02	149.96	6	1	400
MR04	20.02	149.96	6	3	300
MR04	20.02	149.96	6	6	200
MR04	20.02	149.96	6	19	120
MR04	20.02	149.96	6	21	70
MR04	20.02	149.96	6	23	10
M35	32.99	155.27	17	1	5000
M35	32.99	155.27	17	8	4000
M35	32.99	155.27	17	13	2800
M35	32.99	155.27	16	2	2000
M35	32.99	155.27	16	10	1000
M35	32.99	155.27	16	11	800
M35	32.99	155.27	16	13	650
M35	32.99	155.27	16	15	500
M35	32.99	155.27	16	17	300
M35	32.99	155.27	16	19	200
M35	32.99	155.27	16	21	50
M35	32.99	155.27	16	24	10
M30	28.47	154.82	2	1	5800
M30	28.47	154.82	2	4	5000
M30	28.47	154.82	2	7	4000
M30	28.47	154.82	2	13	3000
M30	28.47	154.82	2	15	2000
M30	28.47	154.82	9	1	1000
M30	28.47	154.82	9	5	800
M30	28.47	154.82	9	7	700
M30	28.47	154.82	9	9	500
M30	28.47	154.82	9	11	400
M30	28.47	154.82	9	13	270
M30	28.47	154.82	9	16	120

M30	28.47	154.82	9	19	50
M30	28.47	154.82	9	21	10
M22	20.02	154.97	6	1	5600
M22	20.02	154.97	6	11	4000
M22	20.02	154.97	6	17	3000
M22	20.02	154.97	6	24	2000
M22	20.02	154.97	10	8	1000
M22	20.02	154.97	10	9	800
M22	20.02	154.97	10	11	600
M22	20.02	154.97	10	13	500
M22	20.02	154.97	10	15	400
M22	20.02	154.97	10	18	250
M22	20.02	154.97	10	19	120
M22	20.02	154.97	10	21	50
M22	20.02	154.97	10	23	10
M18	16.00	155.00	5	4	4900
M18	16.00	155.00	5	11	3000
M18	16.00	155.00	5	14	2000
M18	16.00	155.00	9	2	1000
M18	16.00	155.00	9	5	800
M18	16.00	155.00	9	7	600
M18	16.00	155.00	9	10	430
M18	16.00	155.00	9	12	400
M18	16.00	155.00	9	14	300
M18	16.00	155.00	9	18	150
M18	16.00	155.00	9	20	70
M18	16.00	155.00	9	22	10
K8a	12.51	154.94	4	1	5900
K8a	12.51	154.94	4	13	4000
K8a	12.51	154.94	4	18	3000
K8a	12.51	154.94	4	23	2000
K8a	12.51	154.94	9	1	1000
K8a	12.51	154.94	9	4	800
K8a	12.51	154.94	9	6	600
K8a	12.51	154.94	9	8	400
K8a	12.51	154.94	9	11	300
K8a	12.51	154.94	9	13	250
K8a	12.51	154.94	9	16	150
K8a	12.51	154.94	9	19	90
K8a	12.51	154.94	9	21	50
K8a	12.51	154.94	9	22	10
K13	10.98	129.96	6	1	5650

K13	10.98	129.96	6	2	5000
K13	10.98	129.96	6	3	4500
K13	10.98	129.96	6	4	4000
K13	10.98	129.96	6	6	3000
K13	10.98	129.96	6	7	2500
K13	10.98	129.96	6	8	2000
K13	10.98	129.96	6	9	1500
K13	10.98	129.96	6	10	1000
K13	10.98	129.96	6	11	800
K13	10.98	129.96	6	12	600
K13	10.98	129.96	6	13	500
K13	10.98	129.96	6	14	300
K13	10.98	129.96	6	15	200
K13	10.98	129.96	6	17	150
K13	10.98	129.96	6	19	110
K13	10.98	129.96	6	23	50

#### References

Bowman, K. and C.R. Hammerschmidt (2011) Extraction of monomethylmercury from seawater for low-femtomolar determination, Limnol. Oceanogr. Methods, 9, 121-128.68 Gill, G.A. and W.F. Fitzgerald (1985) Mercury sampling of open ocean waters at the

picomolar level, Deep-Sea Research, 32, 287-297.

Peltzer, E.T. and Brewer, P.G. (1993) Some practical aspects of measuring DOC — sampling artifacts and analytical problems with marine samples Marine Chemistry 41(1-3), 243-252.

#### 4.5.3. Dissolved Inorganic Carbon (DIC)

Samples for DIC were collected with 12-liter Niskin bottles mounted onto a Rosette sampling assembly, equipped with a CTD recorder (Sea-Bird SBE911 plus), at all stations with full depth profiles. Samples for DIC analysis were overflowed by at least one bottle volume, stored in 250 mL PYREX<sup>®</sup> borosilicate glass bottles, and poisoned with 50  $\mu$ L of HgCl<sub>2</sub>-saturated solution upon sample collection. The DIC concentration will be measured by acidifying ~0.5 mL of water sample and subsequently quantifying released CO<sub>2</sub> using an infrared CO<sub>2</sub> detector (Apollo SciTech model AS-C3) with a precision of ±2  $\mu$ mol kg<sup>-1</sup> (Cai et al., 2004). DIC will be calibrated against certified reference material provided by Dr. A. G. Dickson at the Scripps Institution of Oceanography, University of California, San Diego.

#### References

Cai, W.-J., M. Dai, Y. Wang, W. Zhai, T. Huang, S. Chen, F. Zhang, Z. Chen and Z. Wang, 2004. The Biogeochemistry of Inorganic Carbon and Nutrients in the Pearl River Estuary and the Adjacent Northern South China Sea, Continental Shelf Research, 24: 1301-1319.

#### 4.5.4. Dissolved oxygen (DO)

Duplicate samples for DO were collected from 12-liter Niskin bottles mounted onto a Rosette

sampling assembly, equipped with a CTD recorder (Sea-Bird SBE911 plus), at all stations with full depth profiles. The DO concentration in discrete samples were measured on board within ~24 hours using the spectrophotometric Winkler method (Labasque et al., 2004; Pai et al., 1993), with a precision of  $\pm 1 \mu$ mol kg<sup>-1</sup>.Samples for DO were collected into BOD bottles with a volume of ~60 mL and overflowed by at least one bottle volume, avoiding the introduction of bubbles. Immediately after seawater subsampling, 0.5 mL of MnCl<sub>2</sub> solution and 0.5 mL of NaI-NaOH solution were successively added to the sample bottles. The sample bottles were capped and shook upside down at least 20 times to fully fix the DO in seawater as MnO(OH)<sub>2</sub> precipitate. After incubation in a 25±0.02 °C water bath for several hours to allow for precipitates to settle down to the bottle, ~0.5 mL of 28% (v/v) H<sub>2</sub>SO<sub>4</sub> was added to the precipitation to release I<sub>2</sub>. The I<sub>2</sub> concentrations were then measured spectrophotometrically at 466 nm with the spectrophotometer (Model UV-1800, Shimadzu Suzhou Instruments, Manufacturing Co., Ltd.) and calibrated by standard curves using KIO<sub>3</sub> standard solutions.

#### References

- Labasque, T., C. Chaumery, A. Aminot and G. Kergoat, 2004. Spectrophotometric Winkler determination of dissolved oxygen: Re-examination of critical factors and reliability, Marine Chemistry, 88: 53-60.
- Pai, S.-C., G.-C. Gong and K.-K. Liu, 1993. Determination of dissolved oxygen in seawater by direct spectrophotometry of total iodine, Marine Chemistry, 41: 343-351.

#### 4.5.5. Total Alkalinity (TA)

Samples for TA were collected with 12-liter Niskin bottles mounted onto a Rosette sampling assembly, equipped with a CTD recorder (Sea-Bird SBE911 plus), at all stations with full depth profiles. Samples for TA analysis are overflowed by at least one bottle volume, stored in 250 mL PYREX<sup>®</sup> borosilicate glass bottles, and poisoned with 50  $\mu$ L of a HgCl<sub>2</sub>-saturated solution upon sample collection. TA will be determined by a Gran titration with hydrochloric acid using an automated Alkalinity Titrator (Apollo SciTech model AS-ALK1+) with a precision of better than ±2  $\mu$ mol kg<sup>-1</sup>. TA will be calibrated against certified reference material provided by Dr. A. G. Dickson at the Scripps Institution of Oceanography, University of California, San Diego.

#### 4.5.6. pH

Samples for pH were collected with 12-liter Niskin bottles mounted onto a Rosette sampling assembly, equipped with a CTD recorder (Sea-Bird SBE911 plus), at all stations with full depth profiles. Samples for pH analysis were overflowed by at least one bottle volume, stored in 250 mL PYREX<sup>®</sup> borosilicate glass bottles, and poisoned with 50  $\mu$ L of HgCl<sub>2</sub>-saturated solution upon sample collection. pH will be measured at 25±0.01 °C via UV-VIS spectrophotometer (Agilent 8453) using the unpurified meta-cresol purple (mCP) from Acros Origanics on the total hydrogen ion concentration scale (pH<sub>T</sub>) in the seawater. The precision of analysis is better than ±0.0005 pH units (Dickson et al., 2007).

#### References

Dickson, A., S. Chris and J. R. Christian, 2007. Guide to Best Practices for Ocean CO2

Measurements, Guide to Best Practices for Ocean CO<sub>2</sub> Measurements, PICES Special Publication 3191 pp..

#### 4.5.7. Partial pressure of carbon dioxide (pCO<sub>2</sub>)

The surface seawater partial pressure of carbon dioxide was continuously measured onboard with a cavity ring-down analyzer (Picarro G2301) integrated into an underway automated flowing  $pCO_2$  system (Model 8050, General Oceanic Inc. USA). The schematic diagram of the underway  $pCO_2$  system is shown in Fig. 9. The system is highly compact and operates by directing seawater to flow through a chamber (the equilibrator) where the CO<sub>2</sub> contained in the seawater equilibrates with the gas in the chamber (the headspace gas). The equilibrated gas in the headspace is pumped into a cavity ring-down analyzer (Picarro G2301) to measure its CO<sub>2</sub> mole fraction ( $xCO_2$ ) instantaneously, and then returned to the equilibrator, forming a closed loop. Periodically, atmospheric air is also pumped through the analyzer to measure its CO<sub>2</sub> mole fraction. The analyzer is calibrated with four CO<sub>2</sub> standard gases at regular intervals. Meanwhile, temperature and salinity of flowing seawater are measured by temperature and salinity sensors (Seabird SBE21), respectively, and dissolved oxygen is measured by oxygen sensor (Aanderaa 4330) throughout the system for analytical, troubleshooting, and quality control purposes.



Figure 5. Schematic diagram of the underway pCO<sub>2</sub> system

#### 4.5.8. Total Thorium-234 (<sup>234</sup>Th)

4-L samples for total <sup>234</sup>Th were collected into FLPE bottles with 12-liter Niskin bottles mounted onto a Rosette sampling assembly, equipped with a CTD recorder (Sea-Bird SBE911 plus), at all stations with full depth profiles. <sup>234</sup>Th is co-precipitated with MnO<sub>2</sub> and filtered through a QMA filter (25 mm, 1.0 μm). All samples for total and particulate <sup>234</sup>Th are dried and mounted on plastic discs with two layers of aluminum foil and one layer of Mylar film. <sup>234</sup>Th is counted for at least 12 h until 2500 counts is obtained using a gas-flow proportional low-level RISØ beta counter. A second count will be carried out after 6 months later to get the background

value. The <sup>234</sup>Th recovery is monitored by adding ~10 dpm <sup>230</sup>Th and analyzed using the demounted total <sup>234</sup>Th samples from the <sup>230</sup>Th spike on QMA filters after beta counting (see details in Cai et al., 2015). The <sup>230</sup>Th is monitored using <sup>229</sup>Th, purified using iron precipitation and anion column exchange, and finally diluted in 2% (v/v) HNO<sub>3</sub>. The ratios of <sup>229</sup>Th and <sup>230</sup>Th were measured with Inductively Couple Plasma-Mass Spectrometry (ICP-MS).

#### 4.5.9. Particulate Organic Carbon (POC)

8-L samples for POC were collected into FLPE bottles with 12-liter Niskin bottles mounted onto a Rosette sampling assembly, equipped with a CTD recorder (Sea-Bird SBE911 plus) at all stations within the upper 1000 m. Generally, 8L samples were filtered onto pre-combusted (450 °C, 5 hours) Whatman QMA filters (25 mm, 1  $\mu$ m) at pressure of 0.3 atm. The filter was dried overnight at 50 °C and firstly used for the measurements of <sup>234</sup>Th. Prior to analysis, POC samples will be placed overnight in a desiccator saturated with HCl fumes. The filters will then be dried again at 50 °C and packed in tin-foil film. POC will be analyzed on a Vario EL cube CHNS Elemental Analyzer following the guidelines provided by the manufacturer as dried, acidified samples of particulate matter is combusted at 960 °C and release CO<sub>2</sub> converted from organic carbon and N<sub>2</sub> subsequently reduced from nitrogen oxides. Both gases are measured by thermal conductivity.

#### References

Cai, P., Zhao, D., Wang, L., Huang, B., & Dai, M. (2015). Role of particle stock and phytoplankton community structure in regulating particulate organic carbon export in a large marginal sea. *Journal of Geophysical Research: Oceans*, 120(3), 2063-2095.

#### 4.5.10. Dissolved Barium (DBa) and its isotopes ( $\delta^{138}Ba_{DBa}$ )

Seawater samples for the concentrations and isotopic compositions of DBa were collected with 12-liter Niskin bottles mounted onto a Rosette sampling assembly, equipped with a CTD recorder (Sea-Bird SBE911 plus), at all stations with full depth profiles. Samples were stored in

250 mL polyethylene bottles immediately after sampling and subsequently acidified to pH~2 with

0.1% (v/v) distilled concentrated HCl and stored at room temperature in the dark until analysis in the laboratory. DBa concentrations in seawater will be analyzed using an isotope dilution method (Klinkhammer and Chan, 1990; Freydier et al., 1995) on an Agilent 7500 quadrupole-ICP-MS,

and  $\delta^{138}Ba_{DBa}$  will be determined using double spike technique (Rudge et al., 2009) in the static mode on a Nu Plasma HR MC-ICP-MS.

#### References

- Klinkhammer, G.P., and Chan, L.H. (1990). Determination of barium in marine waters by isotope dilution inductively coupled plasma mass spectrometry. *Analytical Chimica Acta*, 232, 323-329.
- Freydier, R., Dupre, B., and Polve, M. (1995). Analyses by inductively coupled plasma mass spectrometry of Ba concentrations in water and rock samples. Comparison between isotope dilution and external calibration with or without internal standard. *European Mass Spectrometry*. 1(3), 283-291.
- Rudge, J.F., Reynolds, B.C., and Bourdon, B. (2009). The double spike toolbox. *Chemical Geology*. 265, 420-431.

#### 4.5.11. Particulate Barium (PBa) and its isotopes ( $\delta^{138}Ba_{PBa}$ )

Samples for the concentrations and isotopic compositions of PBa were collected with 12-liter Niskin bottles mounted onto a Rosette sampling assembly, equipped with a CTD recorder (Sea-Bird SBE911 plus), at three Mega stations (M35/M22/K8a) with full depth profiles. About 8 L of seawater was filtered through 0.4  $\mu$ m polycarbonate membranes, which were then dried at 50°C overnight and stored in polycarbonate dishes until analysis in the laboratory. The suspended particle samples will be dissolved completely using the digestion method developed by Cardinal et al. (2001) and the resultant digested solution will be dried down and re-dissolved in small amounts of 1 mol L<sup>-1</sup> HCl. Both Ba and Al will be measured using a quadrupole-ICP-MS (Agilent 7500) and PBa concentrations can be calculated by the excess above the lithogenic Ba/Al ratios (Jacquet et al., 2008).  $\delta^{138}$ Ba<sub>PBa</sub> will be determined using double spike technique (Rudge et al., 2009) in the static mode on a Nu Plasma HR MC-ICP-MS.

#### References

- Jacquet, S.H., Dehairs, M.F., Savoye, N., Obernosterer, I., Christaki, U., Monnin, C., and Cardinal, D. (2008). Mesopelagic organic carbon mineralization in the Kerguelen Plateau region tracked by biogenic particulate Ba. *Deep Sea Research*. 55, 868-879.
- Cardinal, D., Dehairs, F., Cattaldo, T., and André, L. (2001). Geochemistry of suspendedparticles in the Subantarctic and Polar Front Zones south of Australia: Constraints of export and advection processes. *Journal of Geophysical Research Biogeosciences*. 106, 31637-31656.

#### 4.5.12. Dissolved silicon isotopes ( $\delta^{30}$ Si<sub>Si(OH)4</sub>)

Seawater samples for dissolved silicon isotopes were collected with 12-liter Niskin bottles mounted onto a Rosette sampling assembly, equipped with a CTD recorder (Sea-Bird SBE911 plus), at all stations with full depth profiles. Samples were stored in 250 mL polyethylene bottles immediately after sampling and subsequently acidified to  $pH\sim2$  with 0.1% (v/v) distilled concentrated HCl and stored at room temperature in the dark until analysis in the laboratory. Si(OH)<sub>4</sub> will be separated from the major matrix elements using a two-step brucite

co-precipitation technique adapted from the MAGIC method (Karl and Tien, 1992). The precipitates will be further purified using cation-exchange chromatography (Georg et al., 2006).  $\delta^{30}Si_{Si(OH)4}$  will be determined in the pseudo high-resolution mode on a Nu Plasma HR MC-ICP-MS.

#### References

- Georg, R.B., Reynolds, B.C., Frank, M., and Halliday, A.N. (2006). New sample preparation techniques for the determination of Si isotopic compositions using MC-ICPMS. *Chemical Geology*. 235, 95–104.
- Karl, D.M., and Tien, G. (1992). MAGIC: A sensitive and precise method for measuring dissolved phosphorus in aquatic environments. *Limnology&Oceanography*. 37, 105–116.

#### 4.5.13. Particulate biogenic silicon (BSi) and its isotopes (δ<sup>30</sup>Si<sub>BSi</sub>)

Samples for the concentrations and isotopic compositions of particulate biogenic silicon were collected with 12-liter Niskin bottles mounted onto a Rosette sampling assembly, equipped with a CTD recorder (Sea-Bird SBE911 plus), by filtering ~8 L of seawater through 0.4  $\mu$ m polycarbonate membranes (at three Mega stations (M35/M22/K8a) ~30 L of seawater were filtered) with full depth profiles. The membranes were then dried at 50°C overnight and stored in polycarbonate dishes until analysis in the laboratory. The suspended particle samples will be dissolved using the double wet-alkaline digestion method (Rgueneau et al., 2005). BSi concentrations will be analyzed with a Technicon AA3 Auto-Analyzer (Bran+Luebbe GmbH) following classical colorimetric methods, and  $\delta^{30}Si_{BSi}$  will be determined in the pseudo high-resolution mode on a Nu Plasma HR MC-ICP-MS.

#### **References:**

Ragueneau, O., Savoye, N., Amo, Y.D., Cotten, J., Tardiveau, B., and Leynaert, A. (2005). A new method for the measurement of biogenic silica in suspended matter of coastal waters: using Si:Al ratios to correct for the mineral interference. *Continental Shelf Research*. 25, 697-710.

#### 4.5.14. 210Po-210Pb

<sup>210</sup>Po-<sup>210</sup>Pb disequilibrium in the wNP will be used to investigate the particle dynamics in the oligotrophic mesopelagic and deep waters and to determine the export fluxes of biogenic particles out of the euphotic zone.

#### **Analytical Methods:**

Dissolved <sup>210</sup>Po-<sup>210</sup>Pb: For operationally defined dissolved Po and Pb, the water samples are filtered through the PC membrane with a pore size of 0.4  $\mu$ m and acidified immediately to pH<2 with reagent grade 6M HCl after sampling onboard. <sup>209</sup>Po spike, stable Pb (PbCl<sub>2</sub>) and FeCl<sub>3</sub> will be added into the filtrate after returning to the onshore laboratory. Pb and Po will be simultaneously co-precipitated with Fe(OH)<sub>3</sub>. The Fe(OH)<sub>3</sub> precipitate will be collected and redissolved in HCl, then <sup>210</sup>Po and <sup>209</sup>Po will be spontaneously deposited onto silver plates for alpha spectrometry analysis.

Particulate <sup>210</sup>Po-<sup>210</sup>Pb: The particulate samples retained on the PC membrane will be dried in an oven and weighed to estimate the concentration of total suspended matter. After adding <sup>209</sup>Po spike, the filters will be digested by mixed concentrated HNO<sub>3</sub>/HCl/HClO<sub>4</sub> solution. <sup>210</sup>Po and <sup>209</sup>Po will be deposited onto silver plates and counted following the same procedures as dissolved phases. All the samples of Particulate and dissolved <sup>210</sup>Po-<sup>210</sup>Pb was collected above 1000m for 10 liters per layer, a total of 12 layers in all clean stations. And full depth for 10 liters per layer, a total of 24 layers in mega stations.

#### 4.5.15. <sup>230</sup>Th, <sup>231</sup>Pa, <sup>232</sup>Th

<sup>230</sup>Th, <sup>231</sup>Pa, <sup>232</sup>Th in seawater will be determined to investigate the particle dynamics, to quantify the lithogenic dust deposition flux, and to help obtaining the residence times of biogenic TEIs in the wNP. The water samples were filtered in-line with capsule filters (0.8 μm/0.45 μm Acropak® 500 filters) and acidified immediately with optima HCl to pH<2 after the sample collection. After weighting, spiking and preconcentration with Mg hydroxide, <sup>230</sup>Th, <sup>232</sup>Th, and <sup>231</sup>Pa will be separated and purified with AG1×8 resin and then measured with Element XR HR-ICP-MS. Sampling strategy and depth of seawater <sup>230</sup>Th, <sup>231</sup>Pa, <sup>232</sup>Th samples are consistent with samples for <sup>210</sup>Po-<sup>210</sup>Pb.

#### 4.5.16. Nd isotopes

The dissolved Nd isotope ratio ( $^{143}$ Nd/ $^{144}$ Nd, expressed as  $\epsilon$ Nd) of seawater is widely used as water mass tracer in the different ocean basins. In our study, sources of macro- and micro-nutrients to the North Pacific Subtropical Gyre from terrestrial input (e.g. island), water mass transport, and hydrothermal vent etc. will be examined using  $\epsilon$ Nd. During this cruise, seawater samples of Nd isotopes from surface to bottom were taken from 12 L Niskin bottles mounted on a Rosette sampler equipped with a calibrated CTD recorder. Upon sampling, 10L or 20 L of seawater were filtered on board in a clean bench with 0.45 µm Polyethersulfone membrane filter into pre-cleaned LDPE cubitainers. The filtrate was acidified to a pH of ~2 with ultra-clean 6 N HCl (self-distilled or Trace metal grade). Nd isotope will be analyzed using MC-ICP-MS in the laboratory on land. Seawater samples were taken from hydro CTD at Stations K2b, WPS, MR04, M35, M30, M22, M18, K8a, and K13 from surface to bottom. The detailed information is presented in the table below.

		1	8	I	
Station	Latitude	Longitude	Cast	Bottle	Depth
Station	(°N)	(°E)	Number	Number	(m)
K2b	20.59	126.22	19	1	5000
K2b	20.59	126.22	19	2	5000
K2b	20.59	126.22	19	3	3000
K2b	20.59	126.22	19	4	3000
K2b	20.59	126.22	19	5	3000
K2b	20.59	126.22	19	6	2000

Table 12 The detailed sampling information of Nd isotopes

K2b	20.59	126.22	19	7	1000
K2b	20.59	126.22	19	8	1000
K2b	20.59	126.22	19	9	800
K2b	20.59	126.22	19	10	800
K2b	20.59	126.22	19	11	640
K2b	20.59	126.22	19	12	640
K2b	20.59	126.22	19	13	500
K2b	20.59	126.22	19	14	500
K2b	20.59	126.22	19	15	350
K2b	20.59	126.22	19	16	350
K2b	20.59	126.22	19	17	170
K2b	20.59	126.22	19	18	170
K2b	20.59	126.22	19	19	100
K2b	20.59	126.22	19	20	100
K2b	20.59	126.22	19	21	50
K2b	20.59	126.22	19	22	50
K2b	20.59	126.22	19	23	10
K2b	20.59	126.22	19	24	10
WPS	19.98	140.00	13	1	4600
WPS	19.98	140.00	13	2	4600
WPS	19.98	140.00	13	3	3000
WPS	19.98	140.00	13	4	3000
WPS	19.98	140.00	13	5	2000
WPS	19.98	140.00	13	6	2000
WPS	19.98	140.00	13	7	1000
WPS	19.98	140.00	13	8	1000
WPS	19.98	140.00	13	9	800
WPS	19.98	140.00	13	10	800
WPS	19.98	140.00	13	11	600
WPS	19.98	140.00	13	12	600
WPS	19.98	140.00	13	13	500
WPS	19.98	140.00	13	14	500
WPS	19.98	140.00	13	15	300
WPS	19.98	140.00	13	16	300
WPS	19.98	140.00	13	17	200
WPS	19.98	140.00	13	18	200
WPS	19.98	140.00	13	19	150
WPS	19.98	140.00	13	20	150
WPS	19.98	140.00	13	21	70
WPS	19.98	140.00	13	22	70
WPS	19.98	140.00	13	23	25
WPS	19.98	140.00	13	24	25
MR04	20.02	149.96	2	1	4600

MR04	20.02	149.96	2	2	4600
MR04	20.02	149.96	2	5	2000
MR04	20.02	149.96	2	6	2000
MR04	20.02	149.96	2	12	1000
MR04	20.02	149.96	2	13	1000
MR04	20.02	149.96	2	16	800
MR04	20.02	149.96	2	17	800
MR04	20.02	149.96	2	20	600
MR04	20.02	149.96	2	21	600
MR04	20.02	149.96	2	23	500
MR04	20.02	149.96	2	24	500
MR04	20.02	149.96	6	1	400
MR04	20.02	149.96	6	2	400
MR04	20.02	149.96	6	3	300
MR04	20.02	149.96	6	4	300
MR04	20.02	149.96	6	5	200
MR04	20.02	149.96	6	6	200
MR04	20.02	149.96	6	19	120
MR04	20.02	149.96	6	20	120
MR04	20.02	149.96	6	21	70
MR04	20.02	149.96	6	22	70
MR04	20.02	149.96	6	23	10
MP04	20.02	149.96	6	24	10
1V1IX04	20.02				
M35	32.99	155.27	16	1	2000
M35 M35	32.99 32.99	155.27 155.27	16 16	1 2	2000 2000
M35 M35 M35	32.99 32.99 32.99	155.27 155.27 155.27	16 16 16	1 2 9	2000 2000 1000
M35 M35 M35 M35 M35	32.99 32.99 32.99 32.99 32.99	155.27 155.27 155.27 155.27	16 16 16 16	1 2 9 10	2000 2000 1000 1000
M35 M35 M35 M35 M35 M35	32.99 32.99 32.99 32.99 32.99 32.99	155.27 155.27 155.27 155.27 155.27	16 16 16 16 16	1 2 9 10 11	2000 2000 1000 1000 800
M35 M35 M35 M35 M35 M35 M35	32.99 32.99 32.99 32.99 32.99 32.99 32.99	155.27 155.27 155.27 155.27 155.27 155.27	16 16 16 16 16 16	1 2 9 10 11 12	2000 2000 1000 1000 800 800
M35 M35 M35 M35 M35 M35 M35 M35	32.99 32.99 32.99 32.99 32.99 32.99 32.99 32.99	155.27 155.27 155.27 155.27 155.27 155.27 155.27	16 16 16 16 16 16 16	1 2 9 10 11 12 13	2000 2000 1000 800 800 650
M35 M35 M35 M35 M35 M35 M35 M35 M35	32.99 32.99 32.99 32.99 32.99 32.99 32.99 32.99 32.99	155.27 155.27 155.27 155.27 155.27 155.27 155.27 155.27	16 16 16 16 16 16 16 16	1 2 9 10 11 12 13 14	2000 2000 1000 1000 800 800 650 650
M35 M35 M35 M35 M35 M35 M35 M35 M35 M35	32.99 32.99 32.99 32.99 32.99 32.99 32.99 32.99 32.99 32.99	155.27 155.27 155.27 155.27 155.27 155.27 155.27 155.27 155.27	16 16 16 16 16 16 16 16 16	1 2 9 10 11 12 13 14 15	2000 2000 1000 800 800 650 650 500
M35 M35 M35 M35 M35 M35 M35 M35 M35 M35	32.99 32.99 32.99 32.99 32.99 32.99 32.99 32.99 32.99 32.99 32.99	155.27 155.27 155.27 155.27 155.27 155.27 155.27 155.27 155.27 155.27	16 16 16 16 16 16 16 16 16 16	1 2 9 10 11 12 13 14 15 16	2000 2000 1000 800 800 650 650 650 500 500
M35 M35 M35 M35 M35 M35 M35 M35 M35 M35	32.99 32.99 32.99 32.99 32.99 32.99 32.99 32.99 32.99 32.99 32.99 32.99 32.99	155.27 155.27 155.27 155.27 155.27 155.27 155.27 155.27 155.27 155.27 155.27	16 16 16 16 16 16 16 16 16 16 16	1 2 9 10 11 12 13 14 15 16 17	2000 2000 1000 800 800 650 650 650 500 500 300
M35 M35 M35 M35 M35 M35 M35 M35 M35 M35	32.99 32.99 32.99 32.99 32.99 32.99 32.99 32.99 32.99 32.99 32.99 32.99 32.99 32.99	155.27 155.27 155.27 155.27 155.27 155.27 155.27 155.27 155.27 155.27 155.27 155.27 155.27	16 16 16 16 16 16 16 16 16 16 16 16	1 2 9 10 11 12 13 14 15 16 17 18	2000 2000 1000 800 800 650 650 650 500 500 300 300
M35 M35 M35 M35 M35 M35 M35 M35 M35 M35	32.99 32.99 32.99 32.99 32.99 32.99 32.99 32.99 32.99 32.99 32.99 32.99 32.99 32.99 32.99 32.99	155.27 155.27 155.27 155.27 155.27 155.27 155.27 155.27 155.27 155.27 155.27 155.27 155.27 155.27	16 16 16 16 16 16 16 16 16 16 16 16 16	1 2 9 10 11 12 13 14 15 16 17 18 19	2000 2000 1000 800 800 650 650 650 500 500 300 300 200
M35 M35 M35 M35 M35 M35 M35 M35 M35 M35	32.99 32.99 32.99 32.99 32.99 32.99 32.99 32.99 32.99 32.99 32.99 32.99 32.99 32.99 32.99 32.99 32.99	155.27 155.27 155.27 155.27 155.27 155.27 155.27 155.27 155.27 155.27 155.27 155.27 155.27 155.27 155.27 155.27	16 16 16 16 16 16 16 16 16 16 16 16 16 1	1 2 9 10 11 12 13 14 15 16 17 18 19 20	2000 2000 1000 800 800 650 650 650 500 500 500 300 300 200 200
M35 M35 M35 M35 M35 M35 M35 M35 M35 M35	32.99 32.99 32.99 32.99 32.99 32.99 32.99 32.99 32.99 32.99 32.99 32.99 32.99 32.99 32.99 32.99 32.99 32.99 32.99	155.27 155.27 155.27 155.27 155.27 155.27 155.27 155.27 155.27 155.27 155.27 155.27 155.27 155.27 155.27 155.27 155.27	16 16 16 16 16 16 16 16 16 16 16 16 16 1	1 2 9 10 11 12 13 14 15 16 17 18 19 20 21	2000 2000 1000 800 800 650 650 650 500 500 300 300 200 200 200 50
M35 M35 M35 M35 M35 M35 M35 M35 M35 M35	32.99 32.99 32.99 32.99 32.99 32.99 32.99 32.99 32.99 32.99 32.99 32.99 32.99 32.99 32.99 32.99 32.99 32.99 32.99 32.99	155.27 155.27 155.27 155.27 155.27 155.27 155.27 155.27 155.27 155.27 155.27 155.27 155.27 155.27 155.27 155.27 155.27 155.27	16 16 16 16 16 16 16 16 16 16 16 16 16 1	1 2 9 10 11 12 13 14 15 16 17 18 19 20 21 22	2000 2000 1000 800 800 650 650 650 500 500 300 300 200 200 50 50 50
M35 M35 M35 M35 M35 M35 M35 M35 M35 M35	32.99 32.99	155.27 155.27 155.27 155.27 155.27 155.27 155.27 155.27 155.27 155.27 155.27 155.27 155.27 155.27 155.27 155.27 155.27 155.27 155.27	16 16 16 16 16 16 16 16 16 16 16 16 16 1	1 2 9 10 11 12 13 14 15 16 17 18 19 20 21 22 23	2000 2000 1000 800 800 650 650 650 500 500 300 300 200 200 200 50 50 50 10
M35 M35 M35 M35 M35 M35 M35 M35 M35 M35	32.99 32.99	155.27 155.27 155.27 155.27 155.27 155.27 155.27 155.27 155.27 155.27 155.27 155.27 155.27 155.27 155.27 155.27 155.27 155.27 155.27 155.27	16 16 16 16 16 16 16 16 16 16 16 16 16 1	1 2 9 10 11 12 13 14 15 16 17 18 19 20 21 22 23 24	2000 2000 1000 800 800 650 650 650 500 500 500 300 200 200 200 50 50 10 10
M35 M35 M35 M35 M35 M35 M35 M35 M35 M35	32.99 32.99	155.27 155.27 155.27 155.27 155.27 155.27 155.27 155.27 155.27 155.27 155.27 155.27 155.27 155.27 155.27 155.27 155.27 155.27 155.27 155.27	$     \begin{array}{r}       16 \\       17 \\       \end{array} $	1 2 9 10 11 12 13 14 15 16 17 18 19 20 21 22 23 24 1	2000 2000 1000 800 800 650 650 500 500 500 300 200 200 200 50 50 50 10 10 10 5000

M35	32.99	155.27	17	8	4000
M35	32.99	155.27	17	9	4000
M35	32.99	155.27	17	13	2800
M35	32.99	155.27	17	14	2800
M30	28.47	154.82	2	1	5800
M30	28.47	154.82	2	2	5800
M30	28.47	154.82	2	4	5000
M30	28.47	154.82	2	7	4000
M30	28.47	154.82	2	8	4000
M30	28.47	154.82	2	13	3000
M30	28.47	154.82	2	15	2000
M30	28.47	154.82	2	16	2000
M30	28.47	154.82	9	1	1000
M30	28.47	154.82	9	2	1000
M30	28.47	154.82	9	5	800
M30	28.47	154.82	9	6	800
M30	28.47	154.82	9	7	700
M30	28.47	154.82	9	8	700
M30	28.47	154.82	9	9	500
M30	28.47	154.82	9	10	500
M30	28.47	154.82	9	11	400
M30	28.47	154.82	9	12	400
M30	28.47	154.82	9	13	270
M30	28.47	154.82	9	14	270
M30	28.47	154.82	9	16	120
M30	28.47	154.82	9	17	120
M30	28.47	154.82	9	19	50
M30	28.47	154.82	9	20	50
M30	28.47	154.82	9	21	10
M30	28.47	154.82	9	22	10
M22	20.02	154.97	6	1	5600
M22	20.02	154.97	6	2	5600
M22	20.02	154.97	6	11	4000
M22	20.02	154.97	6	12	4000
M22	20.02	154.97	6	17	3000
M22	20.02	154.97	6	18	3000
M22	20.02	154.97	6	23	2000
M22	20.02	154.97	6	24	2000
M22	20.02	154.97	10	7	1000
M22	20.02	154.97	10	8	1000
M22	20.02	154.97	10	9	800
M22	20.02	154.97	10	10	800
M22	20.02	154.97	10	11	600

M22	20.02	154.97	10	12	600
M22	20.02	154.97	10	13	500
M22	20.02	154.97	10	14	500
M22	20.02	154.97	10	15	400
M22	20.02	154.97	10	16	400
M22	20.02	154.97	10	17	250
M22	20.02	154.97	10	18	250
M22	20.02	154.97	10	19	120
M22	20.02	154.97	10	20	120
M22	20.02	154.97	10	21	50
M22	20.02	154.97	10	22	50
M22	20.02	154.97	10	23	10
M22	20.02	154.97	10	24	10
M18	16.00	155.00	5	4	4900
M18	16.00	155.00	5	6	4000
M18	16.00	155.00	5	11	3000
M18	16.00	155.00	5	14	2000
M18	16.00	155.00	9	1	1000
M18	16.00	155.00	9	2	1000
M18	16.00	155.00	9	4	800
M18	16.00	155.00	9	5	800
M18	16.00	155.00	9	6	600
M18	16.00	155.00	9	7	600
M18	16.00	155.00	9	10	430
M18	16.00	155.00	9	11	430
M18	16.00	155.00	9	12	400
M18	16.00	155.00	9	13	400
M18	16.00	155.00	9	14	300
M18	16.00	155.00	9	15	300
M18	16.00	155.00	9	18	150
M18	16.00	155.00	9	19	150
M18	16.00	155.00	9	20	70
M18	16.00	155.00	9	21	70
M18	16.00	155.00	9	22	10
M18	16.00	155.00	9	23	10
K8a	12.51	154.94	4	1	5900
K8a	12.51	154.94	4	2	5900
K8a	12.51	154.94	4	13	4000
K8a	12.51	154.94	4	14	4000
K8a	12.51	154.94	4	18	3000
K8a	12.51	154.94	4	19	3000
K8a	12.51	154.94	4	23	2000
K8a	12.51	154.94	4	24	2000

K8a	12.51	154.94	9	1	1000
K8a	12.51	154.94	9	2	1000
K8a	12.51	154.94	9	4	800
K8a	12.51	154.94	9	5	800
K8a	12.51	154.94	9	6	600
K8a	12.51	154.94	9	7	600
K8a	12.51	154.94	9	8	400
K8a	12.51	154.94	9	9	400
K8a	12.51	154.94	9	10	300
K8a	12.51	154.94	9	11	300
K8a	12.51	154.94	9	12	250
K8a	12.51	154.94	9	13	250
K8a	12.51	154.94	9	15	150
K8a	12.51	154.94	9	16	150
K8a	12.51	154.94	9	18	90
K8a	12.51	154.94	9	19	90
K8a	12.51	154.94	9	20	50
K8a	12.51	154.94	9	21	50
K8a	12.51	154.94	9	22	10
K8a	12.51	154.94	9	23	10
K13	10.98	129.96	6	2	1000
K13	10.98	129.96	6	4	700
K13	10.98	129.96	6	8	400
K13	10.98	129.96	6	9	400
K13	10.98	129.96	6	11	265
K13	10.98	129.96	6	12	265
K13	10.98	129.96	6	14	200
K13	10.98	129.96	6	15	200
K13	10.98	129.96	6	16	130
K13	10.98	129.96	6	17	130
K13	10.98	129.96	6	19	80
K13	10.98	129.96	6	23	10

#### 4.5.17. Hg

For the THg concentration analysis, HCl (conc.) is added to 250 ml seawater sample to form a final concentration of 0.5% and the samples are stored at -20 °C before analysis. Samples will be oxidized with 0.05% bromine monochloride (BrCl) solution or equivalent for at least 1 hour. Then excess halogens will be removed by mixing with 0.05% v/v hydroxylamine hydrochloride (NH<sub>2</sub>OH•HCl) solution for at least 5 minutes, and final reduction will be conducted with 0.05% v/v stannous chloride (SnCl<sub>2</sub>) solution. Hg<sup>0</sup> in solution will be purged and trapped on gold or gold-coated sand (or the equivalent). Purging should proceed at a volumetric flow rate of no more than 1 L min<sup>-1</sup> (we recommend 0.5 L min<sup>-1</sup>) until a volume of gas of at least 15 times the volume

of liquid has been sparged. Then the gold will be detected by AFS.

For MeHg analysis,  $H_2SO_4$  (conc.) is added to 250 ml seawater sample to form a final concentration of 1% and the samples are stored at -20 °C before analysis. Upon arrival at the laboratory, the sample is first neutralized with 6 mL of 50% KOH, and then buffered to pH=5 with 1.8 mL of 2 M Na-Acetate/Acetic buffer, the pH should be checked and adjusted as necessary with small additions of strong acid (H<sub>2</sub>SO<sub>4</sub>) or strong base (KOH). 0.18 mL of 1% NaTEB will be added to the buffered 250 ml sample, allowing each sample to react for at least 15 minutes, and then sparging the formed MeHgEt from the sample to a Tenax trap. MeHgEt on the trap will then be detected by a packed column GC (OV-3)-AFS.

#### 4.6 In-situ Pump (Prof. Dai)

#### 4.6.1. Objectives:

- To determine the particulate concentrations and isotopes of the major elements, e.g. POC, PN and biogenic silica.
- 2) To determine the particulate concentrations and isotopes of the trace elements, e.g. Fe, Zn, Cd, Co, Cu, Ni and REEs.
- 3) To determine the particulate activities of the natural occurring radionuclides, e.g. <sup>234</sup>Th.
- 4) To evaluate the size-fractionated information and particle dynamics of the trace elements.
- 5) To estimate the downward fluxes of the trace elements.

#### 4.6.2. Sample strategy and collection

We re-designed the configuration of the two pump heads in situ pump system based on the GEOTRACES cookbook in order to satisfy the trace element sampling requirements for the GEOTRACES cruise (See Figure 6). Briefly, for one pump head, seawater sequentially passes through a 51 $\mu$ m Nitex (acid cleaned) and 0.8  $\mu$ m PES membrane (acid cleaned) for trace element analysis. For the other pump head, seawater also passes through a 51 $\mu$ m Nitex (acid cleaned) and 1.0  $\mu$ m QMA filter (400 °C combusted) for major element (or natural radionuclides) analysis. One-way valves were used to prevent the back flow of seawater to the pump heads which may induce contamination.



Figure 6 The new design of in-situ pump water transfer system with two pump heads

#### 4.6.3. Sample processing:

- 1/4 51µm Nitex filter and 1/16 0.8µm PES membrane from PES pump head were used for particulate trace element (PTE) analysis. The Nitex filter samples were firstly washed with clean filtered seawater onto a 25mm 0.8µm PES membrane. Both PES membranes were dried under room temperature in a clean flow bench (Class 100).
- 2) 1/4 51µm Nitex filter and one punch of 23 mm diameter QMA filter from QMA pump head were used for POC, PN and particulate <sup>234</sup>Th analysis. The Nitex filter samples were firstly washed onto a 25 mm diameter 1.0m QMA filter. Both QMA filters were mounted on plastic discs with a layer of aluminum foil and mylar cover, and beta counted onboard with a low-level beta counter.
- 1/16 51µm Nitex filter from QMA pump head and 1/16 0.8µm PES membrane from PES pump head were dried under room temperature in a clean flow bench (Class 100) for biogenic silica analysis.
- 1/8 51µm Nitex filter and two punches of 23 mm diameter QMA filters from QMA pump head were dried in an oven at 50 °C for particulate inorganic carbon (PIC) analysis.
- 5) 1/8 51µm Nitex filter and two punches of 23 mm diameter QMA filter from QMA pump head were used for particulate amino acids (PAA) analysis. The Nitex filter samples were firstly washed onto a 25 mm diameter 1.0m QMA filter. Both QMA filters were preserved at -20 °C.

#### 4.6.4. Sampling information:

During this cruise we performed pump deployment at 9 clean stations with 16 casts in total (1-2 casts per station), and 4-7 pumps were deployed during each cast. The deployment depths and

pumping volumes are shown in Table 13.

Station	Cast	Depth <sup>a</sup>	QMA-V <sup>b</sup> [L]	PES-V <sup>b</sup> [L]	Total-V [L]
K2b	1	75	1091.65	517.50	1601.50
K2b	1	130	1233.90	646.90	1878.50
K2b	1	165	1173.00	631.30	1789.00
K2b	1	220	1300.30	618.95	1932.10
K2b	1	330	1190.60	593.95	1777.55
K2b	1	550	1255.05	609.50	1859.00
K2b	2	25	874.10	502.30	1373.50
K2b	2	850	1205.00	537.15	1766.20
K2b	2	1050	1269.15	582.85	1882.35
K2b	2	1650	1139.00	522.00	1655.00
K2b	2	2200	1189.90	559.30	1774.50
K2b	2	3250	1124.05	440.70	1583.50
WPS	1	600	1205.75	597.75	1798.00
WPS	1	1000	1179.70	572.60	1756.30
WPS	1	1600	1165.90	489.35	1852.35
WPS	1	2300	824.00	373.00	1213.00
WPS	1	3000	1190.40	511.85	1725.50
WPS	1	3500	1181.90	401.20	1607.00
WPS	2	30	966.35	522.20	1469.00
WPS	2	80	993.60	529.55	1518.50
WPS	2	140	1088.50	601.00	1687.20
WPS	2	200	1263.50	662.45	1924.65
WPS	2	400	1259.40	617.05	1901.00
WPS	2	500	1065.85	442.00	1508.00
MR04	1	30	936.65	529.50	1463.00
MR04	1	80	841.20	440.75	1270.00
MR04	1	140	1073.30	600.10	1675.65
MR04	1	200	1261.50	665.50	1929.00
MR04	1	400	1160.50	605.00	1760.00
MR04	1	600	1287.00	590.45	1891.50
MR04	1	800	1246.50	514.75	1761.00
M35	1	90	78.30	36.75	115.00
M35	1	140	1146.20	617.55	1760.85
M35	1	200	1232.50	685.35	1916.85
M35	1	320	1231.20	646.25	1903.50
M35	1	450	1220.95	558.75	1778.00
M35	2	45	644.35	341.60	982.00
M35	2	550	211.80	91.50	301.00

Table 13 The deployment depths and pumping volumes of two pump heads

M35	2	750	1192.80	548.95	1779.60
M35	2	1300	1231.00	581.50	1819.70
M35	2	2000	1241.10	520.00	1770.00
M35	2	2500	1208.30	435.10	1651.00
M30	1	300	944.65	470.50	1422.70
M30	1	400	1012.60	499.50	1506.00
M30	1	600	995.95	454.85	1448.00
M30	1	800	1039.50	465.90	1507.45
M30	1	1000	1026.80	456.10	1489.50
M30	1	1200	1029.40	373.35	1403.00
M30	2	30	642.80	365.55	1004.00
M30	2	130	699.95	354.10	1059.20
M30	2	150	749.65	401.90	1151.05
M30	2	200	758.60	330.30	1088.50
M22	1	55	811.45	464.40	1271.00
M22	1	150	893.60	480.30	1377.80
M22	1	180	958.80	508.85	1461.00
M22	1	250	1020.75	514.95	1534.70
M22	1	350	1039.00	476.85	1518.50
M22	1	500	1020.85	399.05	1420.00
M22	2	650	593.95	274.40	867.00
M22	2	800	1262.90	516.85	1774.00
M22	2	1100	1260.90	523.50	1791.00
M22	2	1500	1254.30	494.50	1834.23
M22	2	2000	1292.10	494.50	1794.50
M22	2	2800	1191.50	457.75	1658.50
M18	1	50	663.25	378.70	1039.00
M18	1	100	704.55	383.35	1097.67
M18	1	150	743.50	401.45	1140.00
M18	1	185	747.05	328.15	1074.00
M18	2	300	1005.00	494.20	1495.00
M18	2	400	1029.20	440.30	1464.00
M18	2	650	1074.56	433.65	1506.50
M18	2	900	1050.75	446.20	1507.90
M18	2	1100	1017.05	416.90	1439.40
M18	2	1400	1004.20	396.50	1400.00
K8a	1	50	858.60	479.50	1335.50
K8a	1	140	886.00	507.20	1387.00
K8a	1	200	1013.85	460.90	1478.40
K8a	1	300	1043.70	477.50	1525.50
K8a	1	400	1031.09	479.20	1510.00
K8a	1	500	1037.05	385.30	1421.60

K8a	2	600	1257.10	548.60	1802.50
K8a	2	800	1272.90	507.90	1775.00
K8a	2	1000	1248.85	533.30	1787.30
K8a	2	1500	1253.35	551.15	1826.40
K8a	2	2000	1281.80	518.65	1797.50
K13	1	45	764.95	413.40	1171.20
K13	1	110	787.90	391.55	1173.00
K13	1	150	803.75	404.05	1209.50
K13	1	300	925.50	406.45	1338.90
K13	1	500	910.10	411.20	1320.00
K13	1	600	969.90	322.00	1289.50

Notes: (a) The depth information is determined from the length of winch cable; (b) QMA-V and PES-V refer to the pumping volumes for the QMA and PES pump heads, respectively.

#### 4.7. Aerosol Sampling (Trace Elements and Isotopes, Prof. Zhang)

#### 4.7.1. Objectives:

It has been recognized that atmospheric deposition might be an important source of micro-nutrient (e.g. Fe, Mn, Cu) and macro-nutrient (N, P) to the euphotic zone of open oceans. For example, the seasonal variation of Fe concentration in the upper layer of station ALOHA (time-series station in Hawaii) had significant correlation with the variability of dust deposition. However, the observation data in the North Pacific Subtropical Gyre (NPSG) are scant. In our study, we aim to quantify the micro-nutrient (Fe etc) input from atmospheric deposition (aerosol and rainwater) and reveal the spatial variability and their origin in the NPSG. For the above purposes, we focus on the trace elements (Al, Ti, Fe, V, Zn, Pb, Mn, Ni, Cu, Hg), REEs (La, Ce, Pr, Nd, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb, Lu), Ion ( $SO_4^{2-}$ , Cl<sup>-</sup>) in the atmospheric deposition.

#### 4.7.2. Parameters:

Trace elements (Al, Ti, Fe, V, Zn, Pb, Mn, Ni, Cu, Hg), REEs (La, Ce, Pr, Nd, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb, Lu), major anions (SO<sub>4</sub><sup>2-</sup>, Cl<sup>-</sup>), and nutrients (NO<sub>3</sub><sup>-</sup>, PO<sub>4</sub><sup>3-</sup>).

#### 4.7.3. Sampling Information:

Aerosol samples were collected using a high-volume (1000 L/min) air sampler on the compass deck with Whatman 41 membrane filter (low ash cellulose esters, PN 1441-047) during this cruise. The samples were preserved at -20 degree centigrade on board. Rainwater samples are collected by rainwater sampler and stored in LDPE bottle. In this cruise, 23 aerosol samples were collected with cellulose filter which include 15 underway sampling samples and 8 fixed samples while the number of rainwater samples were 17.

#### 4.8. Biological Oceanography (Profs. Shi, and Liu)

#### 4.8.1. Phytoplankton chlorophyll a, biomass and community structure

Chlorophyll-a concentrations: 500 mL seswater by regular CTD per sample (5m, 25m, 50m, 75m, 100m, DCM, 150m, 200m) was filtered onto Machery Nagel GF/F filter papers and extracted for 16-20 hours in 10 mL 90% analytical reagent acetone in a -20 °C freezer in the dark before measurement on a Turner Designs fluorometer following the method of Welschmeyer (1994).

High Performance Liquid Chromatography (HPLC): 4–6 L seawater per sample by Regular CTD (5m, 25m, 50m, 75m, 100m, DCM, 150m, 200m) was filtered onto Machery Nagel GF/F filter papers and placed directly into a -80 °C freezer. These will be analysed the method of e.g. Gibb et al. (2000). Chlorophyll-a concentrations determined by HPLC will be used to verify those determined by fluorimetry (above).

Analytical flow cytometry: 1.8 mL of seawater was mixed with 20uL 16% paraformaldehyde yielding a final paraformaldehyde concentration of 1%. Samples were mixed by hand and left for 10 minutes at room temperature in the dark before transfer to a  $-80^{\circ}$ C freezer. Samples will be analysed on a FACSort flow cytometer following the method of e.g. Davey et al. (2008), with the intention of analysing for nanophytoplankton, picophytoplankton, and total bacterial cell counts. Water for experiments above was collected at the following dates/times (all Beijing time):

#### **References:**

Nicholas A. Welschmeyer. 1994. Fluorometric analysis of chlorophyll a in the presence of chlorophyll b and pheopigments . Limnol. Oceanogr. 39: 1985-1992, doi : 10.4319/lo.1994.39.8.1985.

Davey, M., Tarran, G. A., Mills, M. M., Ridame, C., Geider, R. J., and La Roche, J. 2008. Nutrient limitation of picophytoplankton photosynthesis and growth in the tropical North Atlantic. Limnol. Oceanogr. 53: 1722–1733, DOI:10.2307/40058292.

Gibb, S. W., Barlow, R. G., Cummings, D. G., Rees, N. W., Trees, C. C., Holligan, P., and Suggett, D. 2000. Surface phytoplankton pigment distributions in the Atlantic Ocean: an assessment of basin scale variability between 50 degrees N and 50 degrees S. Prog. Oceanogr. 45: 339–368, DOI:10.1016/S0079-6611(00)00007-0.

#### 4.8.2. Mesozooplankton biomass and community structure

This cruse carried out UVP5 devolution along with CTD at 9 stations (K2b, M35, M30, WPS, KPR, MR05, K8a, M22 and M26a). The operating depth was the same as that of CTD (200m, 800m, 1000m and full water column). The distribution pattern information and plankton image information of particles with different particle sizes in the water column were obtained.

130 mesozooplankton samples were collected at 8 stations (K2b, M35, M30, WPS, M18, MR04, K8a, And M22) on this voyage. Four trawling operations were conducted at time continuous stations, and two trawling operations were conducted at other key stations, all close to noon or midnight.

In the stance divide five layers (50 m at the bottom of the corresponding mixed layer, corresponding DCM layer at the bottom of the 100 m and 200 m corresponding true light corresponding weak light, 400 m at the bottom of the layer corresponding weak light layer at the bottom of the central, 700 m), using multiple Multinet plankton trawl samples. Motoda sampler was used to divide the collected medium-sized zooplankton on the research ship. Half of the samples were put into 200mL plastic bottles, and 5% neutral formaldehyde solution was added for fixation and preservation. ZOOSCAN was used to analyze and measure the samples in the laboratory. The other half of the sample was frozen in a 200ml plastic bottle for later use.

#### **Reference:**

Anna Schukat, Lena Teuber, Wilhelm Hagen, Norbert Wasmund, Holger Auel,2013.Energetics and carbon budgets of dominant calanoid copepods in the northern Benguela upwelling system.Journal of Experimental Marine Biology and Ecology.442:1-9.

Jessica R. Frost, Anneke Denda, Clive J. Fox, Charles A. Jacoby, Rolf Koppelmann, Morten Holtegaard Nielsen, Marsh J. Youngbluth,2012.Distribution and trophic links of gelatinous zooplankton on Dogger Bank, North Sea.Marine Biology.159(2):239-253.

Guidi L, Chaffron S, Bittner L, et al. Plankton networks driving carbon export in the oligotrophic ocean[J]. Nature, 2016, 532(7600):465-470.

Picheral M, Guidi L, Stemmann L, et al. The Underwater Vision Profiler 5: An advanced instrument for high spatial resolution studies of particle size spectra and zooplankton[J]. Limnology & Oceanography Methods, 2010, 8(5):462-473.

#### 4.8.3. N<sub>2</sub> fixation rate and diazotroph composition

Measurement of N<sub>2</sub> fixation rate and primary production, incubations of near-surface seawater with nutrients (Fe and P) and CO<sub>2</sub> manipulations (By Zuozhu Wen, Wenfang Lin, Ruotong Jiang, Ze Chen, Dalin Shi)

Phytoplankton productivity in most of the low latitude oceans are limited by the availability of bioavailable nitrogen. Nitrogen fixation by diazotrophs converts dinitrogen ( $N_2$ ) gas into ammonia, provides nearly half of the ocean's bioavailable nitrogen (Gruber et al., 2008), and is therefore a key regulator of productivity in these regions (Karl et al., 2012; Böttjer et al., 2016). Despite this importance, an observationally-supported, mechanistic understanding of what controls of  $N_2$  fixer biogeography in the ocean and how the diazotrophs may response to the changes in climate, are lacking.

**Objectives:** 

- 1) To estimate the N<sub>2</sub> fixation rate and the diazotroph composition, and their relationships with the environmental factors.
- To conduct Fe and P manipulations experiments, find the limiting nutrient of N<sub>2</sub> fixation in the NPSG.
- To conduct ocean acidification incubations, reveal how the diazotrophs will response to ocean acidification.

Sampling information and preliminary results

Water samples for the measurements of  $N_2$  fixation rate and diazotroph composition were collected at 17 stations at six depths corresponding to 100%, 50%, 25%, 10%, 1%, and 0.1% of the surface irradiance. Water samples for the measurement of other parameters which including primary production, Chl *a* concentration, particulate organic carbon (POC) and nitrogen (PON) concentrations as well as their natural stable isotopic abundance, were also collected at each station.

A matrix of Fe and P manipulation experiments were conducted at 11 stations under trace metal clean condition. Surface (~1-2 m depth) seawater for the experiments is sampled from a custom-built towed fish delivered to a container laboratory over-pressurized with HEPA-filtered air. 12L of seawater was collected into a 10-L polycarbonate carboys (Nalgene Labware), and then spiked with the following combinations of nutrients/trace metals: P, Fe, and combination of both. Triplicate control carboys with no nutrients added were also collected and incubated for 2 days alongside all nutrient-treated carboys.

Ocean acidification experiments were conducted at 11 stations. Seawater was collected into 10-L polycarbonate carboys as described above. Seawater carbonate chemistry was manipulated by gently bubbling seawater with 0.22  $\mu$ m-filtered air or CO<sub>2</sub>-air mixture generated by CO<sub>2</sub> mixers (Ruihua Instrument & Equipment Ltd). All carboys were then incubated for 2 days in the incubators screening to ~45% of sea surface irradiance.

#### **References:**

Gruber, N. and J.N. Galloway, *An Earth-system perspective of the global nitrogen cycle*. Nature, 2008. **451**(7176): p. 293-296.

Karl, D.M., et al., *Predictable and efficient carbon sequestration in the North Pacific Ocean supported by symbiotic nitrogen fixation*. PNAS, 2012. **109**(6): p. 1842-1849.

Böttjer, D., et al., *Temporal variability of nitrogen fixation and particulate nitrogen export at Station ALOHA*. Limnology and Oceanography, 2016. 62(1): p. 200-216.

## 4.8.4. Carbon Fixation Measurements for Net Primary Production (14C-NPP) and Photosynthesis-Irradiance (P-I) Curves

During the cruise, 24-h <sup>14</sup> C uptake measurements to estimate NPP and 4-6 h P-I curve experiments were both conducted. NPP measurements were conducted with water collected in the CTD or underway water in the morning. With photosynthetically available radiation (PAR) sensors attached to the Seabird CTD, water of 6-8 layers from the surface and from depths corresponding to 50, 23, 12, 4, 1, 0.5, and 0.1% of surface PAR was collected. NPP was measured in an incubator on deck. Sunlight was screened by different combinations of neutral and blue density filters (LEE filters, UK) to simulate submarine irradiances. The seawater samples were placed in 60-mL polycarbonate bottles (two light bottles and one dark bottle for each depth) in a water bath, the temperature of which was maintained with seawater pumped from a depth of 5 m. Carbon fixation was estimated from the uptake of NaH<sup>14</sup>CO<sub>3</sub>, which was added in trace amounts to the incubation bottles (Strickland and Parsons, 1972). After 24 h, the samples were filtered onto

25-mm-diameter PC filters. The filters were processed immediately on board the research vessel. The radioactivity (DPM, disintegrations per minute) on the filters was measured with a Tri-Carb 4810TR liquid scintillation counter after removing residual inorganic carbon by acid fuming overnight and immersing the filters in 4 mL of Ultima Gold scintillation cocktail (Perkin-Elmer, USA) until the filters became transparent. P-I curves were measured 3-5 layers of samples from depths of surface, 50, 12, 1 and 0.1% of surface PAR in the morning. The experiments were conducted with a photosynthetron with metal halide lamps (Xie et al., 2015). The seawater samples were placed in twelve 60-mL polycarbonate bottles in a water bath, the temperature of which was maintained with seawater pumped from a depth of 5 m. After a 4-6 h incubation, the samples were filtered and processed following the procedures described above.

The NPP derived from 24-h incubations was calculated as follows:

$$NPP = \frac{1.05 \times DIC(DPM_L - DPM_D)}{DPM_{tot}}$$

where the DPM<sub>D</sub> (DPM in dark bottle) was subtracted from the DPM<sub>L</sub> (DPM in light bottles) to eliminate the influence of adsorption and dark fixation of carbon. DPM<sub>tot</sub> is the total activity of NaH<sup>14</sup>CO 3 added to the incubation bottle. The activity of a small amount of NaH<sup>14</sup>CO<sub>3</sub> was measured to estimate the value of DPM<sub>tot</sub>. DIC is the dissolved inorganic carbon concentration (g m<sup>-3</sup>), which was estimated from salinity using an empirical equation DIC =  $(9.9647+0.3944\times$ Salinity) (Xie et al., 2015). The factor of 1.05 was used to correct for isotope discrimination between <sup>14</sup>C and <sup>12</sup>C. Photosynthetic parameters were derived from the P-I curves by fitting to the equations (Platt et al., 1980):

$$P^{B} = P_{s}^{B} \left[ 1 - exp\left(-\frac{\alpha I}{P_{s}^{B}}\right) \right] exp\left(-\frac{\beta I}{P_{s}^{B}}\right)$$
$$P_{m}^{B} = P_{s}^{B} \left[\frac{\alpha}{\alpha + \beta}\right] \left[\frac{\beta}{\alpha + \beta}\right]^{\frac{\beta}{\alpha}}$$

here  $P^B$  is the chlorophyll-*a*-normalized photosynthetic rate;  $P_m^B$  is the light-saturated photosynthetic rate;  $\alpha$  is the light-limited slope; and  $\beta$  is the photoinhibition parameter. In the absence of photoinhibition,  $\beta = 0$ , and the equation became (Webb et al., 1974):

$$P^{B} = P_{m}^{B} \left[ 1 - exp\left( -\frac{\alpha I}{P_{m}^{B}} \right) \right]$$

From 4 July to 21 August 2020, a total of 47 times of dilution incubation were conducted, including 24 times of CTD water, and 20 times of underway surface water.

Date	Station	Longitude	Latitude	Project
2020/7/6	K2b	126.3	20	NPP\P-I
2020/7/7	K2b-2	126.3	20	NPP\P-I
2020/7/8	K2b-3	126.3	20	NPP\P-I
2020/7/12	KPR	135	16.5	NPP
2020/7/14	WPS	140	20	NPP

Table 14 CTD sample station for NPP

2020/7/15	WPS-2	140	20	NPP
2020/7/19	MR-04	150	20	NPP\P-I
2020/7/24	M35	155	33	NPP\P-I
2020/7/25	M35-2	155	33	NPP
2020/7/26	M35-3	155	33	NPP\P-I
2020/7/27	M32	155	30.5	NPP\P-I
2020/7/28	M30	155	28.5	NPP\P-I
2020/7/31	M26a	158	24	NPP\P-I
2020/8/2	M22	155	20	NPP\P-I
2020/8/3	M22-2	155	20	NPP\P-I
2020/8/4	M20	155	20	NPP\P-I
2020/8/5	M18	155	16	NPP\P-I
2020/8/7	M16	155	14	NPP\P-I
2020/8/8	K8a	155	12.5	NPP\P-I
2020/8/9	K8a-2	155	12.5	NPP
2020/8/11	K9a	150	13	NPP\P-I
2020/8/14	K11a	140	14	NPP\P-I
2020/8/15	K12a	1351	12.75	NPP\P-I
2020/8/17	K13	130	11	NPP

All the sample station including underway and CTD NPP (B-W) and P-I (P-E) showed as follows:



Figure 7 Sample station during the cruise

### References:

Strickland, J. D. H., and Parsons, T. R. (1972). A Practical Handbook of Seawater Analysis.

Ottawa, ON: Fisheries Research Board of Canada.

- Xie, Y. Y., Huang, B. Q., Lin, L. Z., Laws, E. A., Wang, L., Shang, S. L., et al. (2015). Photosynthetic parameters in the northern South China Sea in relation to phytoplankton community structure. J. Geophys. Res. Oceans 120, 4187–4204. doi: 10.1002/2014jc010415
- Platt, T., Gallegos, C. L., and Harrison, W. G. (1980). Photoinhibition of photosynthesis in natural assemblages of marine phytoplankton. J. Mar. Res. 38, 687–701.
- Webb, W. L., Newton, M., and Starr, D. (1974). Carbon dioxide exchange of Alnus rubra: a mathematical model. Occologia 17, 281–291. doi: 10.1007/BF00345747

#### 4.8.5. Phytoplankton growth rate and microzooplankton grazing rate

The growth rate of phytoplankton and the grazing rate of microzooplankton in this experiment were researched by dilution method (Landry & Hassett, 1982). Collecting surface layer and DCM layer water through CTD of the route stations, and collecting surface water between route stations into PC bucket. The non-particle seawater was prepared by gravity filtration using a 0.2 um diameter membrane capsule filter. The non-particle seawater was first injected into 2.4L incubation flask according to the proportion of natural seawater 16.7%, 37.5%, 62.5% and 100%, each dilution gradient set two parallel, then slowly fill the natural seawater culture bottle with silicone tube, tighten the cap, up and down slowly invert several times to make the incubation water mixture; The incubation bottle containing surface flowing water, and the incubation bottle containing DCM layer seawater was placed in the laboratory. All treatments were completed within 1 hour after water extraction.

Before incubation, one sample of chlorophyll a (0.7-2um, 2-20um, >20 um), two samples of pico-phytoplankton (FCM) and one sample of lugols' microscopic examination were collected. At the end of incubation, samples of chlorophyll a with three particles, two samples of pico-phytoplankton (FCM) and lugols' microscopic examination were collected from each incubation bottle. The fractionated chlorophyll a samples were filtered, extracted and determined by Turner Trilogy Fluorimetry, and one sample of FCM was determined by Cytosub on the boat, and the remaining samples were taken back to the laboratory using FACSAria for analysis. The lugols' samples were brought back to the laboratory, they were deposited for 24 hours using a Utermohl device (Utermohl, 1958). The condensed water samples were subjected to an inverted microscope (LEICA DMIRB) with a magnification of 200 to 400X. Microzooplankton species were identified, counted and photographed.

Sample Information

From 4 July to 21 August 2020, a total of 47 times of dilution incubation were conducted, including 24 times of CTD surface layer and DCM layer water, and 23 times of navigation surface water

	Data		Submission
Sampling Indexs	Number	Current State	Date

Table 15 sample information of dilution experiments

ECM	643	Determined and to be submitted	2021.03
FUM	643	Determined and to be submitted	2021.03
	643	Determined and to be submitted	2021.03
Chl a	643	Determined and to be submitted	2021.03
	643	Determined and to be submitted	2021.03
Lugols'	643	Not determine completely	2021.06

#### References

Landry M R, Hassett R P. Estimating the grazing impact of marine microzooplankton[J]. Marine Biology, 1982, 67(3): 283-288

Landry M R, Selph K E, Taylor A G, et al. Phytoplankton growth, grazing and production balances in the HNLC equatorial Pacific[J]. Deep Sea Research Part II: Topical Studies in Oceanography, 2011, 58(3): 524-535

Utermöhl H. Toward the improvement of the quantitative phytoplankton method[J]. Mitteilungen-Internationale Vereiningung für Limnologie, 1958, 9: 1-38

Yumiko Obayashi, Eiichiro Tanoue. Growth and mortality rates of phytoplankton in the northwestern North Pacific estimated by the dilution method and HPLC pigment analysis[J]. Journal of Experimental Marine Biology and Ecology, 2002, 280: 33– 52

#### 4.8.6. Respiration rates

Size fractionated respiration was estimated based on in vitro INT reduction rates as described by E. Elena et al (2019). The sampling depths were 8 layers in the euphotic zone. Four 100 mL polypropylene plastic bottles were filled with seawater. One bottle was immediately fixed by adding formaldehyde (2% final concentration) as a blank. Fifteen minutes later, the four replicates were inoculated in the dark by the addition of 2-para (iodophenyl)-3(nitrophenyl)-5(phenyl) tetrazolium chloride tetrazolium salt (INT) at a final concentration of 0.8 mM. The INT samples were incubated at the in situ temperature. After incubations of 1.5 h, the reactions were stopped by adding formaldehyde. All the samples were sequentially filtered after 15 min through 0.8 and 0.2 µm pore size polycarbonate filters and stored frozen until further processing. The INTf reduction rates were measured using a SP 8001 UV/Vis Spectrophotometer at 485 nm following the procedure of E. Elena et al (2019).

#### **References:**

E. Elena García-Martín, María Aranguren-Gassis, Karl, D. M., Sandra Martínez-García, & Teira,E. 2019. Validation of the in vivo iodo-nitro-tetrazolium (int) salt reduction method as a proxy for plankton respirationdata. Frontiers in Marine ence, 6.