# **Original Research Paper**

# Variability of Organic Matter Sources and Phytoplankton Community Structure During the 19th Century under Global Warming Background in the Chukchi Sea

Zhao Qingying\*, Chen Ronghua, Zhang Haisheng, Yu Peisong and Lu Bing Second Institute of Oceanography, SOA, Hangzhou-310012, China

\*Corresponding author: Zhao Qingying

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# ABSTRACT

Multi-biomarker of sediment core P7200 obtained from the Chukchi Sea during the First Chinese National Arctic Research Expedition was measured to indicate changes of the source of organic matter, the primary productivity and the phytoplankton community structure through the 19th century. The results show that the organic matters in the Chukchi Sea are mainly from the terrigenous source, marine organisms and icerafted debris, and the ratio of terrigenous and oceanic inputs has increased over the 19th century. The total primary productivity has been elevated by nearly 100% in the century and the phytoplankton community structure also exhibits significant variability. The relative abundance of diatom increases dramatically, while that of cocolith decreases obviously and almost constant for the dinoflagellate. It is suggested that the changes of the source of organic matter, the primary productivity and the altered phytoplankton community structure may strengthen the ability of  $CO_2$  absorbing in the Chukchi Sea, which may weaken the greenhouse effect and should be regarded as one of the negative feedback mechanisms to the global warming.

# INTRODUCTION

Since the 20<sup>th</sup> century, human beings have faced a series of severe environmental problems, and global warming has aroused global attention. Carbon dioxide content of the ocean is 55 times more than that of the atmosphere, and marine cycle is controlled by "biological pump". Marine primary producers and their community structure are of important significance to marine ecosystem development and marine "biological pump" process. Climate warming brings significant impact, especially in polar ocean (Screen et al. 2010). The Arctic sea ice is melting quickly; since large volume of freshwater and nutritive salt is injected, the Arctic Pole becomes more sensitive to climate warming and resulting ecosystem changes (Chen et al. 2004, Moritz et al.1996, Manabe et al. 2000, Cuffey et al. 1995). As a primary producer and food web foundation in the Arctic Pole, marine phytoplankton is subject to environmental impact more sensitively and more directly than other organic matters. As a primary producer in marine ecosystem of the Arctic Pole, phytoplankton has become an irreplaceable and important research object during key problem research on changes of the Arctic Ocean (Stroeve et al. 2004). The Arctic Pole can provide early climate records and display global changes with amplifying signals.

To research the response and feedback of the Arctic Pole marine phytoplankton community structure changes to climate warming, investigations and observations with long time series are required. However, we lack historical data about long-term dynamic changes of phytoplankton; it is difficult to obtain data because of very complicated climatic system, cold weather and harsh investigation environment in the Arctic Pole. To cope up with this new challenge, this paper tries to record and reestablish phytoplankton community structure changes in a special way of molecular stratigraphy. There are many successful cases in the international community (Schubert et al. 1998, Werne et al. 2000, Schulte et al. 2003, Higginson et al. 2004, Calvo et al. 2004, Dahl et al. 2004), as well as in China. Zhao et al. (2006, 2009) used 3 specific biomarkers to reestablish phytoplankton productivity and community structure changes in different seas; Zhang et al. (2008) and Xing et al. (2009) used biomarkers to reestablish phytoplankton productivity and community structure change trend in the East China Sea and the Yellow Sea, which were consistent with field investigation data. Bai et al. (2010) and Yu et al. (2014) used 3 specific biomarkers in the Arctic Pole and the South Pole as surrogate markers of phytoplankton community, and successfully reestablished productivity changes.

In the sea of Arctic Pole, phytoplankton, such as diatom, coccolithophores and dinoflagellates, are important primary producers. The content changes of sterol and ketene biomarkers, including brassica sterol (diatom), dinoflagellates sterol (dinoflagellates) and C37 long-chain ketene (coccolithophores), are used to reflect alga structure and primary productivity of specific alga.

At present, planktonic algae represented by diatoms, dinoflagellates and coccolithophores are main contributors to primary productivity of oceans and their total content is regarded to be a surrogate marker of primary productivity. Cholesterol (zooplankter marker) reflects total primary productivity to some extent-flourishing phytoplankton brings rich food to zooplankter and stimulates their growth. By combining these markers, this paper obtains community structure changes of organic matters, understands the impact of past climate warming on marine phytoplankton (specific population) in the Arctic Pole or variability to global warming, and thus provides some valuable illustrations for us. So far, there are no relevant reports about the connection among phytoplankton community structure changes, global warming and marine carbon cycle, which is reflected by 3 alga biomarkers.

# MATERIALS AND METHODS

Chukchi Sea is a marginal sea of the Arctic Ocean. It connects Bering Strait in the south, Chukchi Plateau and Canada Basin in the north. Chukchi Sea is special since it is the only way for the Pacific water to enter the Arctic Ocean. After crossing Bering Strait, the Pacific water is divided into three parts and flows to the Arctic Ocean: low-temperature hyperhaline Anand current in the west, high-temperature hypohaline Alaska coastal current in the east, and the Bering Sea continental shelf water with intermediate nature (Reimnitz et al. 1994, Shi et al. 2004). Meanwhile, Siberia coastal current enters Chukchi Sea through long



Fig. 1: The position of sediment core P7200 from Chukchi Sea.

gorge (Weingartner et al. 2005). Chukchi Sea owns sedimentation of snowfield or polar region, while terrigenous sediment accumulation dominates. 170°W section is a natural line between east part and west part of Chukchi Sea. The Pacific Sea water goes through the Bering Sea and this section and enters the Arctic Ocean. It strengthens biological and biochemical action of sediments along Chukchi Sea and other seas, especially concentration of organic matters.

**Sample source:** Sediment core P7200 is a multi-tube sample obtained from Chukchi Sea during the First Chinese National Arctic Research Expedition in 1999. Research vessel was Snow Dragon; sediment core was 28cm long and water depth was 45m. With uniform lithology, the sample mainly consisted of ash black silt clay and argillaceous silt. Sampling position was 72°00'N, 168°40'W (Fig. 1). Seen from fine particle sediment distribution of the sediment core, Chukchi Sea is characterized by relatively weak hydrodynamic condition and relatively stable sedimentary environment.

Analysis of rate and age of deposition: Deposition rate analysis of multi-tube sample was carried out in State Key Laboratory of Estuarine and Coastal Research, East China Normal University. The sediment was dried at a temperature lower than 40°C, crushed into powder and weighed. The sample was put in a box for 7-10 days and then analyzed; box cover was sealed with wax (Zheng et al. 1983). The instrument was GWL-120210 high-purity germanium coaxial well-type r detecting instrument, produced by the USA ORTE Company. According to measurement with <sup>210</sup>Pbex method, deposition rate and deposition flux of sediment core P7200 was 0.03-0.13cm/a<sup>-1</sup> and 0.28-1.11g·cm<sup>-2</sup>·a<sup>-1</sup> respectively (Figs. 2a-b), which was 0.08cm/a<sup>-1</sup> and 0.69 g·cm<sup>-2</sup>·a<sup>-1</sup> on average. This average value of deposition rate was consistent with 0.7mm/a and 0.89mm/a, measured by Roberts (1995) and Huh (1997) in Greenland northeast sea and in the west of the Arctic Ocean. Stratigraphic time span of sediment core P7200 was 106a (the year 1893-1999).

Lipoid biomarker test pretreatment and analysis: The sediment went through freeze drying. Grind and fetch appropriate quantity of sample, add the mixed solution of dichloromethane and methyl alcohol (3:1) to internal standard; obtain extracting liquid of all layers after 72h Soxhlet extraction, and get dried by rotary evaporation. This concentrated solution went through saponification and hydrolysis (3mL 6% KOH methanol solution), and maintained for 2h at 80°C, in order to get rid of acid components. Normal hexane was used for 4 vibratory extractions; after rotary evaporation, total supernate was concentrated to 0.5mL (protected by nitrogen) and set aside overnight. On the next day, column chromatography was carried out on silica gel: (1)



Fig. 2a-b: The rate and age of deposition of sediment core P7200 from Chukchi Sea.



Fig. 3: The GC atlas of n-alkanes in sediment core P7200 from Chukchi Sea.



Fig. 4: The GC atlas of alcohols and ketene components in sediment core P7200 from Chukchi Sea.

Use normal hexane leaching to obtain alkane component, dry it with nitrogen, move it into 1mL graduated tube and carry out GC analysis on the instrument directly. Adopt HP5890 gas chromatograph (GC) and elastic quartz capillary column (DB-5, 30m×0.25mm inner diameter, 0.17µm coating thickness). Warming procedure: initial temperature was 80°C, warming rate was 5°C/min and finishing temperature was 280°C which was kept for 30 min.

(2) Use more than 10mL normal hexane/dichloromethane (7:3) (add 5% methyl alcohol) to leach and collect silanol and ketene components. Dry it with nitrogen and concen-

trate; alcohols and ketene components needed derivatization reaction. Add dichloromethane and derivatization reagent BSTFA (Bis (trimethylsilyl) trifluoroacetamide to carry out silylanization reaction). After 1h heating reaction, transform to trimethylsilyl ether derivative (TMS-ether), and carry out GC analysis (Agilent 6890N). Capillary-column chromatography was 50m × 0.32mm × 0.17µm; inlet temperature was 300°C and FID detector temperature was 300°C. Adopt unsplit stream sampling; carrier gas was hydrogen whose flow rate was 1.2mL/min. Molecular mark compound was qualitatively determined according to retention time of the



Fig. 5: The content change of biomarkers in sediment core P7200.

spectral peak and quantitatively determined with internal standard method; the content was determined according to area ratio of target peak and internal standard peak (Fig. 4). Use internal standard matter for examination. Recovery rate of this method was >85%; standard deviation of four parallel experiment results was <1%.

#### **RESULTS AND DISCUSSION**

Changes of n-alkanes molecular assemblies, terrigenous source/marine source: Generally speaking, total quantity of alkane in marine sediment is controlled by two factors: alkane production in higher plant source region and environmental conditions to transport it to the ocean; both are mainly subjected to climatic and environmental changes. Terrigenous matters are transported to the ocean in two ways (Hu et al. 2003): direct river input (Bird et al. 1995, Pelejero et al. 1999) and wind transport (Gagosian et al. 1981, Huang et al.1993). It is the main way that long-chain n-alkanes produced by land higher plants are transported into the ocean, settle down and are stored in the sediment (He et al. 2008). Characteristic parameter, specific value and carbon

isotope of n-alkanes can distinguish the source of organic matters in marine sediment to some extent (Meyers et al. 1993, Ratnayake et al. 2006). In general, short-chain n-alkanes without carbon predominance  $(n-C_{15}\sim n-C_{20})$  mainly come from lower fungi and algae in the ocean (Cranwell et al.1987), medium-chain n-alkanes  $(n-C_{20}\sim n-C_{25})$  mainly come from submerged plants, emergent aquatic plants and floating aquatic macrophyte (Ficken et al. 2000, Meyers et al. 2003), long-chain n-alkanes  $(n-C_{27}\sim n-C_{33})$  with obvious odd-carbon number predominance mainly come from land higher plants. Terrigenous plant boasts obvious odd-carbon number predominance, CPI > 5, while CPI of alkanes from oil is close to 1 (Schefu et al. 2003).

Distribution range of n-alkanes in sediment core P7200 is  $nC_{15} \sim C_{33}$ ; main peak carbon in low carbon group is  $C_{17} \sim C_{10}$ and main peak in high carbon group is C<sub>27</sub> or C<sub>29</sub>. Most chromatograms have bimodal pattern (Fig. 3a), showing that organic matters come from terrigenous source and marine source.  $\Sigma C_{15-23}/\Sigma C_{24-33}$  specific value represents the contributions of lower fungi, algae and terrestrial plants respectively. This paper also adopts  $\Sigma$ Odd ( $nC_{25}$ - $nC_{33}$ ) to calculate the total quantity of alkane, as a surrogate marker of terrigenous matter input. The total quantity of alkane is 1242~2010ng/g, CPI is 1.28~2.48, short-chain alkane distribution is relatively even, which may be determined by phytoplankton type of lower fungi and algae. Variation range of ACL is 27.71-28.89, average value is 28.02 and the change is relatively stable at all layers. Pr/Ph specific value is 0.55~1.08; sedimentary environment shows strong reduction-weak oxidation. According to above analysis, organic matters in the sediment core of Chukchi Sea come from 3 sources: terrigenous source, marine organisms and ice-rafted debris (Darby et al. 2009). Moreover, there is one type of special submerged plant, such as watermifoil and pondweed. Its alkane is mainly distributed in  $nC_{21} \sim nC_{25}$ , and main carbon peak is  $nC_{23}$  (Fig. 3b), which are obviously different from terrestrial plant (Ficken et al. 2000, Mwyers et al. 2003).

Chukchi Sea is located in high latitude area, with severe cold climate and widespread sea ice. A lot of terrigenous matters go to every corner of the sea with sea ice; in particular, terrigenous matters from Alaska and north Canada are transported into the sea with ocean current. The most obvious terrigenous effect is that kaolinite content decreases progressively from west to east (Qiu et al. 2007); therefore, it owns sedimentation characteristics of snowfield or polar region. However, affected by warm Pacific water entering through Bering Strait, biological and biochemical action is strengthened during sedimentation. Plankton in the open region of Chukchi Sea is mostly provided by Pacific algae and dissolved substances, whereas terrigenous (foreign) organic

Layer (cm)	Age (a)	$\frac{\Sigma \text{Odd}(n\text{C}_{25}\text{-}n\text{C}_{33})}{(\text{ng/g})}$	ACL	nC <sub>31</sub> /nC <sub>17</sub>	ΣC <sub>15-23</sub> / ΣC <sub>24-33</sub>	СРІ	Pr/Ph	Pr/nC <sub>17</sub>	Ph/nC <sub>18</sub>
0-1cm	1999~	1845	28.24	2.12	0.25	2.00	1.05	0.74	0.85
1-2cm	1999~199 1	2010	28.89	1.67	0.34	3.04	0.78	0.60	1.06
2-3cm	1991~198 0	1784	27.92	2.04	0.45	2.05	0,67	0.93	0.96
3-4cm	1980~196 7	1325	27.55	2.10	0.63	1.11	0.88	0.44	0.79
4-5cm	1967~195 0	1614	27.50	1.54	0.55	2.13	0.55	0.72	1.03
5-6cm	1950~193 0	1242	27.71	0.98	0.80	1.28	0.96	1.05	0.63
6-7cm	1930~189 3	1758	28.66	1.76	0.67	2.05	0.73	0.82	0.59
7-8cm	1893~	1422	27.95	1.05	0.46	1.27	0.49	0.73	0.91

Table 1: Geochemical parameters of n-alkanes in recent sediments from the Chukchi sea.

Note:  $\Sigma Odd$  ( $nC_{25}$ - $nC_{33}$ ) calculates total quantity of alkane; low/high carbon molecule ratio:  $[\Sigma C_{15-23}/\Sigma C_{24-33}]$ ; pristane/ phytane: pr/ph; carbon preference index: CPI= CPI=1/2 [( $\Sigma (C_{25} \sim C_{33})/\Sigma (C_{24} \sim C_{32})$ ]+  $[\Sigma C_{25} \sim C_{33})/\Sigma (C_{26} \sim C_{34})]$ ; average chain length index:

 $ACL = \frac{[nC_{25}] \times 25 + [nC_{27}] \times 27 + [nC_{29}] \times 29 + [nC_{31}] \times 31 + [nC_{33}] \times 33}{[nC_{25}] + [nC_{27}] + [nC_{29}] + [nC_{31}] + [nC_{33}]}$ 

matters are distributed near estuary regions of Chukchi Sea and Alaska large rivers in the form of plant debris.

Relation between biomarker content and plankton community change: Changes in the content and total content of biomarkers in sediment core P7200 are shown in Fig. 5, including brassicasterol (diatom), dinoflagellates sterol (dinoflagellates) and  $C_{37}$  long-chain ketene (coccolithophores). In sediment core P7200, content of brassicasterol is the highest, whereas the contents of dinoflagellates sterol and long-chain ketene are relatively low. In addition, obvious competition between diatom and dinoflagellates indicates that their living environment is relatively similar, so they compete to take in nutrients, which is consistent with researches and modern observation results in this region (Chen et al. 2001, Yang et al. 2002). During the First Chinese National Arctic Research Expedition, among identified phytoplankton, there are 28 genera and 94 species of diatom with high abundance, as well as 2 genera and 6 species of dinoflagellates; average cell density ratio of phytoplankton in Chukchi Sea is 8.32×107 m<sup>-</sup> <sup>3</sup>(Chen et al. 2003). Brassicasterol content changes in 40~446ng/g, dinoflagellates sterol changes in 32~299ng/g, and  $C_{37}$  long-chain ketene changes in 22~230ng/g. In the past century, 4 biomarkers were on the rise on the whole, and their contents changed consistently, represented with x-y linear relation. Fig. 6 shows that correlation between

 $C_{30}$  diol and dinoflagellates sterol is the best ( $R^2 = 0.90$ ); correlation coefficient of  $C_{37}$  long-chain ketene and dinoflagellates sterol is  $R^2 = 0.80$ , that of brassicasterol and dinoflagellates is  $R^2 = 0.85$ . These good correlations show that all biomarkers in sediment core P7200, that is, community changes of marine plants are basically consistent.

Cholesterol is mainly produced by all kinds of marine zooplankton; cholesterol changes in 95~805ng/g in sediment core P7200. Cholesterol content change is basically consistent with dinoflagellates sterol and brassicasterol changes in the last 100 years, but cholesterol content reduces dramatically in the past 30 years, meaning that zooplankton is reducing. Correlation between total content of floating algae biomarkers and cholesterol is  $R^2 = 0.79$ . Main zooplankton in Chukchi Sea is copepods, krills, Chaetognatha and Ostracoda (Chen et al. 2003), they may be the source of cholesterol. Chukchi Sea is a channel connecting North Pacific and the Arctic Ocean. Its ecosystem structure, productivity and marine channel are affected by ring current and dynamic process of sea ice in the Arctic Ocean, and they are also related with the nature of North Pacific water entering Chukchi Sea. In addition, sea ice coverage is an important constraint of primary productivity in the Arctic Ocean.

Relationship between structural changes in pankton

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Fig. 6: The relationship between biomarkers in sediment core P7200.

primary productivity and global warming: Marine phytoplankton community structure is an important factor to control the efficiency of biological pump (Boyd et al.1995,1999), so research about community structure change is of important significance to find climatic change rules. Biomarker content not only serves as a parameter of one phytoplankton content, and their relative percentage composition in total productivity also serves as an index to measure the contribution of every phytoplankton community to total productivity, and reflect the changes of marine phytoplankton community structure. In this paper, three main algae biomarkers: brassicasterol (diatom) + dinoflagellates sterol (dinoflagellates) +  $C_{37}$  long-chain ketene (coccolithophores) = approximate total primary productivity of the ocean. As a whole, respectively calculate the proportions of 3 other biomarkers, as their proportions in total primary productivity of the ocean. This proportion cannot directly reflect real composition of productive group, but its change can reflect relative changes in the contribution of phytoplankton community to production rate. Fig. 7 shows changes in the ratio of main floating algae (diatom, dinoflagellates and coccolithophores) biomarkers to their total content (the total is 100%). It is the change in relative contribution of diatom, dinoflagellates and coccolithophores in the past century; to some extent, this ratio can estimate

changes in phytoplankton community structure. Total primary productivity of biomarkers (brassicasterol, dinoflagellates sterol and coccolithophores) recorded by sediment core P7200 is increased obviously. Relative proportion of diatom increases, dinoflagellates do not fluctuate obviously, and relative content of coccolithophores reduces, which means that 3 phytoplankton community structures have changed obviously in the past century. Diatom and coccolithophores in geologic record play a different role in carbon cycle: diatoms only produce organic carbon, while coccolithophores not only produce organic carbon through photosynthesis, but also produce bones with calcium carbonate, affects marine carbonate and  $CO_2$  system through specific carbonate counter pump, and exerts an important impact on global  $CO_2$  circulation.

In the past several decades, ecosystem of the Arctic Ocean has changed obviously due to global warming (He et al. 2012). In particular, the climate of the North Pole has changed significantly in the last 30 years; sea ice area has shrunk by 5% (Chapman et al.1993). In the Arctic Ocean, water temperature rises, sea ice melts and light transmittance of sea water increases, which helps photosynthesis; phytoplankton and algal bloom and their productivity is further enhanced. In winter, sea ice covers most part of Chukchi Sea and extends to Bering Strait, so variation ranges



Fig. 7: The ratio of three main phytoplankton biomarkers to their total content in sediment core P7200.

of water column temperature and salinity are very narrow (Woodgate et al. 2005). In late spring and summer, sea ice melts; surface water with rich nutritive salts is subject to continuous illumination, and causes brief but strong phytoplankton propagation. During the propagation period, net primary production rate reaches  $300g \cdot cm^{-2} \cdot a^{-1}$  (Walsh et al. 2005). According to estimations with 3 biomarkers in sediment core P7200: brassicasterol (diatom) + dinoflagellates sterol (dinoflagellates) + C<sub>37</sub> long-chain ketene (coccolithophores), total primary productivity of the ocean increases dramatically, which further indicates climatic changes of the North Pole.

Through photosynthesis, marine phytoplankton transforms  $CO_2$  to organic carbon. As one member of phytoplankton, through biological pump process and photosynthesis, coccolithophores changes Ca,  $CO_2$  and water into corresponding quantity of oxygen, organic matters and calcium carbonate. It exerts important influence on carbon cycle of marine ecosystem, effectively restricts global greenhouse effect, and purifies atmospheric environment, hydrosphere and biosphere. This special physiological ecological function of coccolithophores exerts important influence on global air temperature, because coccolith on its cell surface has unique optical property and affects light permeability of sea water like a small mirror. Statistics show that coccolithophores water bloom increases about 6.2%~9.7% marine reflectivity, which weakens global 0.35W/m<sup>2</sup> average radiation intensity and contributes to global temperature reduction significantly(Tyrrell et al.1999). Dimethylsulfide (DMS) produced by coccolithophores is 100 times as much as that of diatom and other algae except *Phaeocystis globosa* (Keller et al.1989), while DMS is a main source of cloud condensation nuclei. By producing DMS, coccolithophores change cloud reflectivity and further affects global air temperature. Reduced coccolithophores in the last 100 years certainly will exert important effects on CO, cycle in the North Pole.

Warming North Pole and increased freshwater runoff bring more freshwater species to the Arctic Ocean; surface sea ice melts. About 40% freshwater is retained in melting ponds on the ice surface, in the ice and under the ice (Eickenet et al. 2002). Therefore, abundance and variety of ice algae have reduced; dominance of freshwater green algae has increased in sea ice and ice-water interface. Previously common zooplankton (nematodes, copepods and amphipoda) have gradually disappeared in the whole ice core (Melnikov et al. 2002). Moreover, increased ultraviolet radiation affects the entire micro and small plankton community structure, and then affects the entire food chain structure (Wickham & Carstens 1998). Maybe, this is the reason why zooplankton in the Chukchi Sea of the North Pole has reduced in recent decades.

#### CONCLUSION

- 1. According to n-alkanes carbon number range, mostly bimodal pattern of main peak carbon and chromatogram, changes of  $\Sigma C_{15\cdot23}/\Sigma C_{24\cdot33}$  ratio,  $\Sigma Odd (nC_{25}-nC_{33})$  of total alkane, carbon preference index (CPI) and average chain length (ACL) of sediment core P7200, it is shown that organic matters in sediments in Chukchi Sea mainly come from terrigenous source, marine organisms and icerafted debris. Moreover, there is one special type of submerged plant, such as watermifoil and pondweed whose alkane is mainly distributed in  $nC_{21} \sim nC_{25}$ , and main carbon peak is  $nC_{23}$ , which are obviously different from terrestrial plants.
- 2. Biomarker records in sediment core P7200 are used to re-establish phytoplankton community structure and its primary productivity changes in the last century. Researches show that estimated by 3 biomarkersbrassicasterol (diatom) + dinoflagellates sterol (dinoflagellates) +  $C_{37}$  long-chain ketene (coccolitho-

phores), total primary productivity of the sea has increased sharply in the last 100 years. The content of dinoflagellates has not fluctuated obviously; relative content of diatoms has risen obviously; and relative content of coccolithophores has dropped. It means that sea and climate of the North Pole have changed, and marine ecology has responded to surrounding climatic changes. As one member of phytoplankton, coccoliths play an important role in carbon cycle of marine ecosystem through biological pump, and also affects seawater carbonate and CO<sub>2</sub> system through special carbonate counter pump. Reduced coccolithophores in the last 100 years certainly will exert important effects on CO<sub>2</sub> cycle in the North Pole (the world).

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