Why are some marginal seas sources of atmospheric CO_2?

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Received 11 March 2013; accepted 18 March 2013; published 29 May 2013.

[1] The contemporary coastal ocean, characterized by abundant nutrients and high primary productivity, is generally seen as a significant CO_2 sink at the global scale. However, mechanistic understanding of the coastal ocean carbon cycle remains limited, leading to the unanswered question of why some coastal systems are sources while others are sinks of atmospheric CO_2. Here we proposed a distinct physical-biogeochemical setting, Ocean-dominated Margin (OceMar), in order for better shaping the concept of the coastal ocean carbon study. OceMars, in contrast to previously recognized River-dominated Ocean Margins, are characterized by dynamic interactions with the open ocean, which may provide nonlocal CO_2 sources thereby modulating the CO_2 fluxes in OceMars. Using the basin areas of the largest marginal seas of the Pacific and the Atlantic, the South China Sea and the Caribbean Sea as examples of OceMars, we demonstrated that such external CO_2 sources controlled the CO_2 fluxes. **Citation:** Dai, M., Z. Cao, X. Guo, W. Zhai, Z. Liu, Z. Yin, Y. Xu, J. Gan, J. Hu, and C. Du (2013), Why are some marginal seas sources of atmospheric CO_2?, *Geophys. Res. Lett.*, *40*, 2154–2158, doi:10.1002/grl.50390.

1. Introduction

[2] Coastal ocean carbon cycling is an important component of the Earth's climate system [*Cai and Dai*, 2004; *Thomas et al*., 2004; *Borges*, 2005; *Borges et al*., 2005; *Cai et al*., 2006; *Chen and Borges*, 2009; *Laruelle et al*., 2010; *Liu et al*., 2010; *Borges*, 2011; *Cai*, 2011; *Lee et al*., 2011]. During the past several years, estimates of global coastal ocean sea-air CO_2 fluxes have dramatically improved as a result of the rapid increase in regional flux measurements. Compilation of the available data sets along with different lines of physical-biogeochemical provinces and/or domains has converged leading to the conclusion that the coastal ocean is an atmospheric sink of ~0.2 to 0.4 Pg C yr^{-1} [*Borges*, 2005; *Borges et al*., 2005; *Cai et al*., 2006; *Chen and Borges*, 2009; *Laruelle et al*., 2010; *Liu et al*., 2010; *Borges*, 2011; *Cai*, 2011].

[3] However, the coastal ocean, or continental margin [*Liu et al*., 2010], remains undersampled both as individual systems for further improved spatial and temporal coverage and at the global scale for better global coverage [*Chen and Borges*, 2009; *Cai*, 2011]. Our current knowledge of CO_2 flux in the coastal ocean is still insufficient to derive precise information for climate change prediction (i.e., flux within $\pm$0.05 Pg C yr^{-1}; [*Cai*, 2011]). More importantly, we still lack a mechanistic understanding as to why some coastal ocean systems act as sinks of atmospheric CO_2 while others are sources. Studies suggest a latitudinal trend of carbon sources or sinks, whereby the low latitude coastal ocean outgases CO_2, due probably to high temperature and high terrestrial organic carbon (OC) input [*Cai and Dai*, 2004; *Borges*, 2005; *Cai et al*., 2006; *Cai*, 2011]. Indeed, low latitude coastal systems are characterized by high temperature and thus higher partial pressure of CO_2 (pCO_2), and recent estimates of global riverine fluxes of dissolved OC (DOC) have again confirmed that up to 60% of riverine DOC export is concentrated in the low latitudinal band of 30°S to 30°N [*Dai et al*., 2012].

[4] However, such a latitudinal trend may not fully withstand close examination of the global pattern of CO_2 fluxes in individual coastal ocean settings. First of all, this trend in the strength of CO_2 sinks lacks a clear indication of temperature or terrestrial OC loading controls. Secondly, there are source and/or near equilibrium regions at higher latitudes. For example, the Scotian shelf acts as a CO_2 source [*Shadwick et al*., 2010] and the English Channel is in near equilibrium with atmospheric CO_2 [*Borges and Frankignoulle*, 2003] although they are both located at 40–60°N. Even in the lower latitudinal systems that are overall sources of atmospheric CO_2, seasonal changes and even reversal from source to sink occur. The northern Gulf of Mexico off the Mississippi River is a moderate source area in the early spring and fall but a moderate sink in summer [*Lohrenz et al*., 2010]. It is thus clear that neither the sink nor the source terms at the seasonal scale can be fully explained by either the thermodynamic change of pCO_2 due to warming/cooling of surface waters, or the degradation of organic matter.

[5] This leads then to a fundamental unresolved question namely, from where is the CO_2 sourced in those continental margins being atmospheric CO_2 sources? An easy hint to approach such a question is to look at upwelling systems. It is interesting that even high latitude continental margins, such as the southern Bering Sea slope north of 50°N influenced by upwelling, may be a strong CO_2 source [*Fransson et al*., 2006]. This strongly suggests that organic material and its decomposition cannot be the sole source

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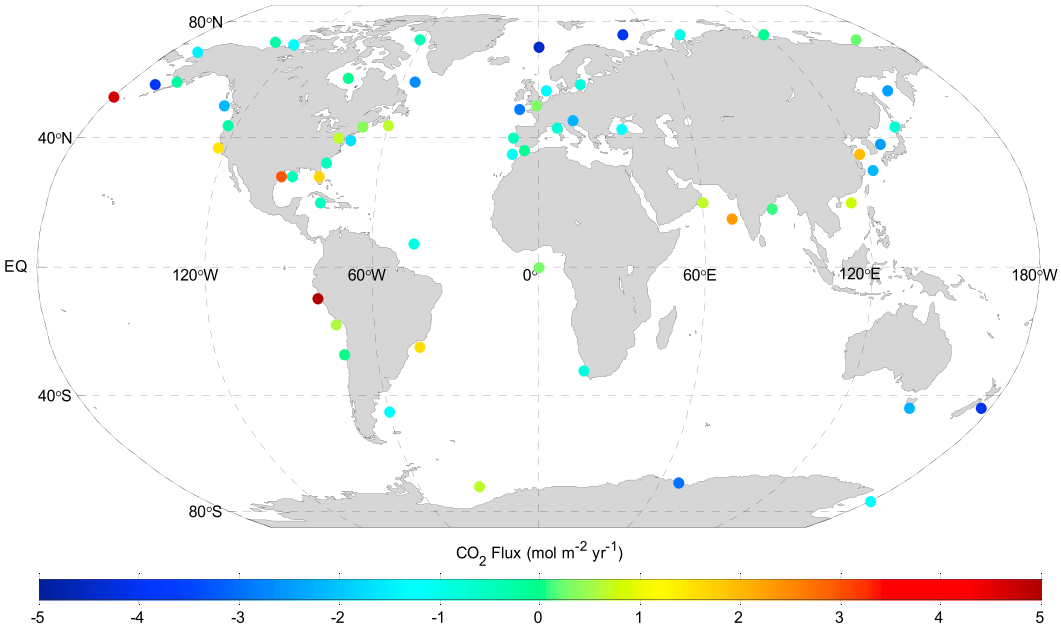

Figure 1. Updated sea-air CO_2 fluxes in the world's coastal oceans. The dataset used in the present estimation is detailed in Table A1.

for the CO_2 degassing. Rather, there should be off-site CO_2 transport which impacts on the regional carbon budget.

2. Hypothesis of Ocean-dominated Margin (OceMar)

[6] In this context, we proposed a new hypothesis: in addition to the processes identical to those in the open ocean such as thermodynamic and biological pump controls, both land input and exchange with the open ocean are significant determinants of the CO_2 fluxes in the coastal ocean. There are at least two distinct settings: River-dominated Ocean Margin (RiOMar; [*McKee et al*., 2004]) and OceMar. In a simplified scheme, RiOMar, as recognized previously, is featured by concurrent inputs of autotrophic (nutrients) and heterotrophic (organic matter) loadings from terrestrial sources at the surface, while OceMar is characterized by concurrent off-site inputs, typically from depth, of nutrients and dissolved inorganic carbon (DIC).

[7] A number of RiOMar systems adjacent to the world's large rivers have been extensively studied and have all shown drawdown of sea surface pCO_2 or significant CO_2 sinks at high river discharges despite the input of organic matter, suggesting high primary productivity and significant net DIC consumption. Examples of such RiOMars include the plumes of the Amazon [*Cooley et al*., 2007], Changjiang [*Tseng et al*., 2011], Pearl [*Cao et al*., 2011], and Mississippi [*Lohrenz et al*., 2010; *Guo et al*., 2012] Rivers (Auxiliary Material). All these RiOMars are characterized by strong seasonality along with river discharge, and the CO_2 fluxes in this type of system are determined by the community metabolism, which is further governed by the competition between heterotrophic and autotrophic loadings.

[8] In contrast, OceMars are characterized by exchange with the open ocean via a two-dimensional (at least) process, i.e., the horizontal intrusion of open ocean water masses when the baroclinic pressure gradient favors their transport into the continental margins, and the subsequent vertical mixing and upwelling. Depending on the characteristics of the source water masses, notably the different ratios of DIC and nutrients into the continental margins, the relative consumption or removal between DIC and nutrients, when being transported into the upper euphotic zones where biogeochemical processes take over, determines the CO_2 fluxes. Under these conditions, excess DIC relative to nutrients existing in the upper layer will eventually lead to CO_2 degassing.

3. Results and Discussion

3.1. Updated Sea-air CO_2 Fluxes in the World's Coastal Oceans

[9] On the basis of the previous syntheses [*Borges et al*., 2005; *Cai et al*., 2006; *Chen and Borges*, 2009; *Laruelle et al*., 2010], we further assessed sea-air CO_2 fluxes in various coastal oceans by including the most up-to-date flux measurements (Figure 1; Table A1). The CO_2 sink in the global coastal ocean is updated to 0.36 Pg C yr^{-1} (Table A1), confirming that the coastal ocean plays a disproportionally important role, comprising 21% of the global ocean net sea-air CO_2 flux of -1.4 Pg C yr^{-1} [*Takahashi et al*., 2009], while accounting for only 7% of its surface area.

3.2. OceMar Case I - The South China Sea (SCS)

[10] We used the northern basin area of the SCS, the largest continental margin of the Pacific, as an OceMar case to examine the CO_2 flux controls. This area is an overall source of atmospheric CO_2 despite seasonal changes [*Zhai et al*., 2005; *Tseng et al*., 2007]. Data were collected in summer 2009 and spring 2011 (Figure A1).

[11] The SCS is semi-enclosed and holds only one source of deep water from the adjacent western North Pacific through the Luzon Strait, where the deepest sill is around 2400 m [*Cao and Dai*, 2011, and references therein]. The deeper part of the SCS is confined to a bowl-shaped trench with the maximum depth around 5560 m, forming a completely isolated basin below

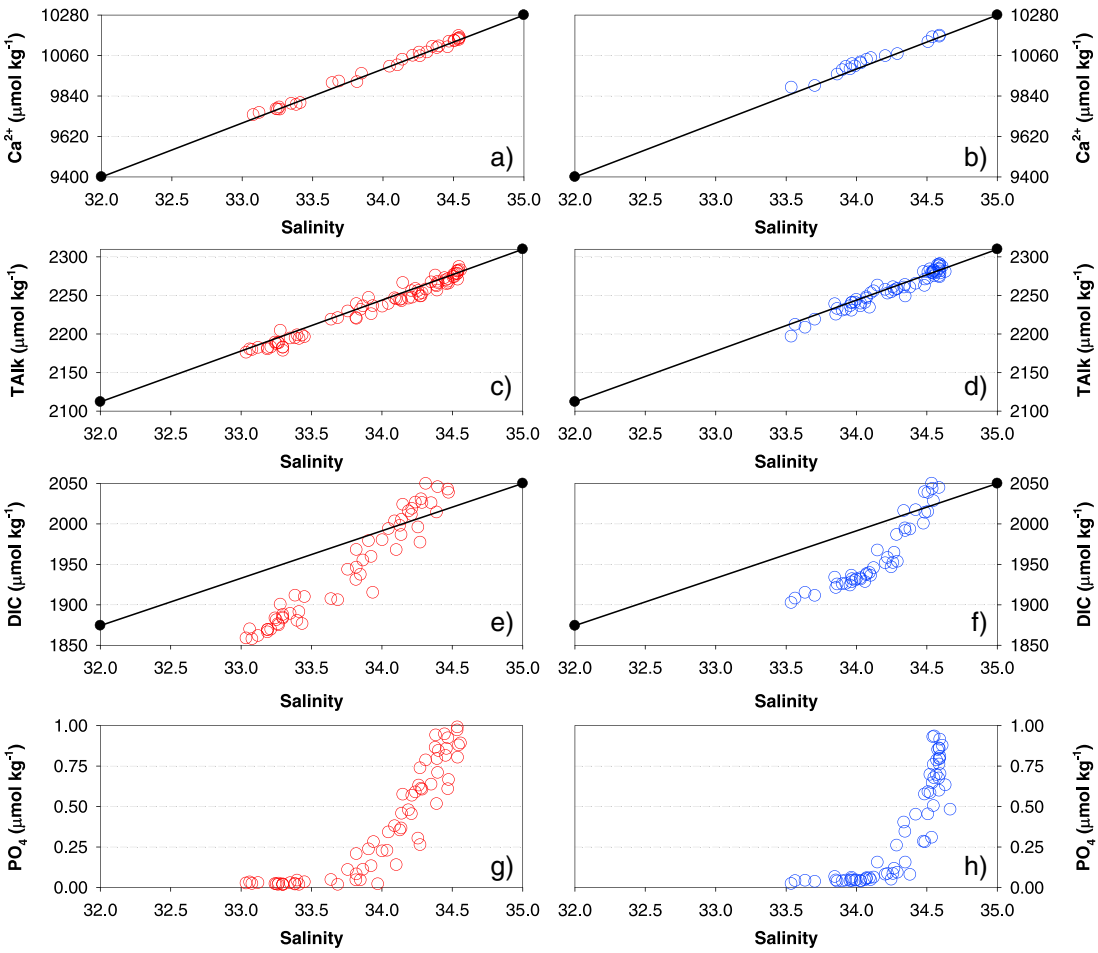

Figure 2. Ca^{2+}, TAlk, DIC, and PO_4 versus salinity in the upper 150 m of the South China Sea basin in (left) summer 2009 and (right) spring 2011. The solid line in panels a–f shows the water mixed by the rain water with zero solutes and the subtropical North Pacific surface water, in which the normalized concentrations of Ca^{2+}, TAlk, and DIC to a salinity of 35 are 10,280 µmol kg^{-1}, 2310 µmol kg^{-1}, and 2050 µmol kg^{-1}, respectively [*Millero*, 1996]. (a) Summer: Ca^{2+} versus salinity; (b) Spring: Ca^{2+} versus salinity; (c) Summer: TAlk versus salinity; (d) Spring: TAlk versus salinity; (e) Summer: DIC versus salinity; (f) Spring: DIC versus salinity; (g) Summer: PO_4 versus salinity; (h) Spring: PO_4 versus salinity.

2400 m. An overflow, driven by a persistent baroclinic pressure gradient, makes the colder and denser Pacific water sink to the deep SCS after it crosses the Luzon Strait [*Wang et al*., 2011, and references therein]. Given the fast vertical replenishment, the deep SCS water must rise to the upper layer through strong diapycnal mixing [*Tian et al*., 2009].

[12] As a result of the unique deep water inflow, we could easily constrain the one-dimensional advection-diffusion model (1-D A/D model; Auxiliary Material) showing that this deep source water of high DIC and phosphate (PO_4) is predominant in the basin area (Figure A2). The deep water-sourced DIC and nutrients when being transported into the upper layer experience biogeochemical alterations, the fraction of which could be estimated as the difference between the predicted values based on conservative mixing between end-members and the measured values. To do so, we first utilized dissolved calcium ions (Ca^{2+}) and total alkalinity (TAlk) as conservative tracers to build the water mass mixing scheme near the surface. Nearly all Ca^{2+} and TAlk samples collected in the upper 150 m of the SCS basin gave values which agreed well with those predicted using a two end-member mixing between the rain water with zero solutes and the subtropical North Pacific surface water (Figure 2a–2d), suggesting that waters in the upper layer of the SCS basin essentially originated from the adjacent open ocean followed by freshwater dilution. In contrast to conservative behavior, DIC showed significant removal at lower salinity during both seasons (Figure 2e and 2f), requiring the nearly complete consumption of PO_4 (Figure 2g and 2h). Such removal in the upper waters of the SCS basin should proceed exclusively through OC production accompanied by nutrient consumption.

[13] In this context, the nonconservative portion of DIC and PO_4 denoted here as ΔDIC and ΔPO_4 in the surface mixed layer of the SCS basin could then be estimated (Auxiliary Material), while the coupling of DIC and nutrient dynamics could be examined based on the classic Redfield ratio of C:N:P = 106:16:1 [*Redfield et al*., 1963]. When ΔDIC exceeded the corresponding $106\Delta PO_4$, extra DIC would be removed beyond the OC production and such "excess ΔDIC" could only be achieved by CO_2 degassing into the atmosphere. In contrast, lower ΔDIC than the corresponding $106\Delta PO_4$ suggested that "deficient ΔDIC" was supplied via the atmospheric CO_2 input to the ocean.

[14] As shown in Figure 3, differences between ΔDIC and $106\Delta PO_4$ in the surface mixed layer of the SCS basin were overall above zero. The average value of $\Delta DIC - 106\Delta PO_4$

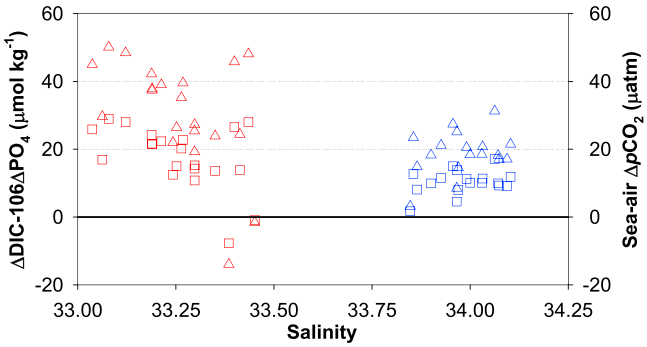

Figure 3. (Squares) $\Delta DIC - 106\Delta PO_4$ and (triangles) sea-air ΔpCO_2 versus salinity in the surface mixed layer of the South China Sea basin in (red symbols) summer 2009 and (blue symbols) spring 2011. The value of 106 is the Redfield C/P uptake ratio (approximately 106/1; [*Redfield et al.*, 1963]). The solid line indicates the pCO_2 equilibrium between the seawater and the atmosphere.

from all stations was $18 \pm 9\,\mu mol\,kg^{-1}$ in summer and $10 \pm 4\,\mu mol\,kg^{-1}$ in spring. Combined with the Revelle factor (Auxiliary Material), the average sea-air pCO_2 difference (ΔpCO_2) resulting from the excess ΔDIC was estimated to be 31 ± 16 μatm in summer and 19 ± 6 μatm in spring. The atmospheric pCO_2 was observed to be ~360 μatm in summer and ~380 μatm in spring. The sea surface pCO_2 in the SCS basin was thus estimated to be 391 ± 16 μatm in summer and 399 ± 6 μatm in spring, which agreed rather well with the field measurements in the SCS basin (387 ± 7 μatm for summer and 407 ± 11 μatm for spring). As a consequence, the SCS basin in both seasons must have worked as a source of CO_2 to the atmosphere, while in summer the source term should be more significant than that in spring.

3.3. OceMar Case II - The Caribbean Sea (CS)

[15] The approach described above was also adopted to diagnose the main controlling processes of CO_2 fluxes in another OceMar system, the CS (Auxiliary Material). The CS is the largest marginal sea of the Atlantic and holds a similar overflow from the Atlantic, yet with a much more complex circulation pattern as compared to the SCS ([*Gordon*, 1967; *MacCready et al.*, 1999]; Auxiliary Material). We focused on the Venezuela Basin (VB) of the CS, the deepest basin, which is ~5000 m deep and has an immediate exchange with the Atlantic through multiple passages. Essentially, Atlantic water enters the VB through relatively shallow passages ranging between 475 m (Mona Passage) and 1815 m (Anegada Passage) in depth (Figure A3). We revealed the CS VB to be a source of CO_2 to the atmosphere in both summer and fall, while it was nearly in equilibrium or a weak sink in spring (Auxiliary Material; Table A2). The estimated sea-air ΔpCO_2 resulting from the excess ΔDIC was 4 ± 5 μatm in summer and 11 ± 6 μatm in fall, consistent with the field observations and remote sensing data [*Olsen et al.*, 2004]. In spring, our estimated sea-air ΔpCO_2 was -7 ± 7 μatm, in reasonable agreement with the value of −10 to 0 μatm [*Olsen et al.*, 2004].

4. Concluding Remarks

[16] The above two cases suggested that while the source of DIC was solely determined by mixing and advection from depth in these two OceMars, the ultimate CO_2 source and/or sink term would be dependent on the relative contribution of DIC and nutrients to the upper layer as well as the biogeochemical alteration therein. These two cases have also emphasized that although the classic controls of atmospheric CO_2 through solubility and biological pumps still stand, other factors must be considered. Of these, DIC additions from off-site and its relative biological removal due to nutrient inputs are other key

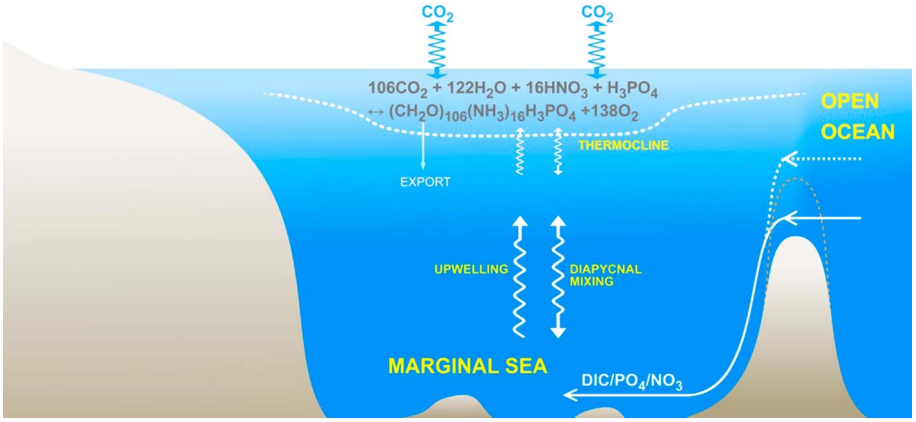

Figure 4. Schematic diagram of the main processes controlling the sea-air CO_2 fluxes in Ocean-dominated Margin (OceMar). For a given marginal sea, the source water mass has a characteristic ratio of DIC and nutrients, which depends on the depth where intrusion from the adjacent open ocean occurs. Such open ocean-originated DIC, PO_4, and nitrate (NO_3) are subsequently transported upward into the thermocline of the marginal sea through vertical mixing and upwelling. The coupled DIC and nutrient consumption via the organic carbon production in the thermocline (or the surface mixed layer) ultimately determines the sea-air CO_2 flux of the marginal sea. Higher ΔDIC than the corresponding $106\Delta PO_4$ or $6.6\Delta NO_3$ suggests that "excess ΔDIC" would be removed by CO_2 degassing into the atmosphere (i.e., CO_2 source), whereas lower ΔDIC than the corresponding $106\Delta PO_4$ or $6.6\Delta NO_3$ suggests that "deficient ΔDIC" would be supplied via the atmospheric CO_2 input to the ocean (i.e., CO_2 sink). The values of 106 and 6.6 are the Redfield C/P and C/N uptake ratios (approximately 106/1 and 106/16; [*Redfield et al.*, 1963]), respectively.

determinants to the CO_2 fluxes. Moreover, although the two cases were both focused on deep basins within marginal seas, the concept of OceMar would be applicable to other ocean margins. For example, the Oregon shelf can be a potential OceMar system where the complete utilization of the preformed nutrients from the upwelled source waters exceeds their corresponding net DIC consumption in the euphotic zone, which may have resulted in the region being a strong CO_2 sink in summer [*Hales et al*., 2005]. We proposed a conceptual model as shown in Figure 4 illustrating the schematic of the major processes in OceMar systems.

[17] The concept of OceMar is to better shape research in the coastal ocean carbon cycle aiming to highlight that in addition to the conventional view of one-dimensional continuity of river-margin-ocean carbon connections, the interaction with the open ocean merits at least two-dimensional examination of physical-biogeochemical processes. Fundamentally, the metabolism of the coastal ocean governed by the relative contribution of DIC, nutrients, and organic material in addition to the physical settings should stand to resolve the carbon mass balance and flux estimation. It should also be pointed out that such classification of RiOMars and OceMars is subject to temporal and spatial changes. A RiOMar system may well be shifted to an OceMar system primarily depending on the physical settings such as circulation and discharges.

[18] **Acknowledgments.** This work was funded by the National Basic Research Program (973) sponsored by the Ministry of Science and Technology through grants 2009CB421200 and 2009CB421201, and the National Natural Science Foundation of China through grants 41121091, 90711005, and 41130857. We thank Yong Zhang, Hua Lin, Xiao Huang, Lifang Wang, Tao Huang, Qian Li, Nan Zheng, Xiuli Yan, Yan Yang, Zongpei Jiang, Liguo Guo, Yuancheng Su, Yanna Wang, Yi Xu, Yan Li, Di Qi, and Zhenyu Sun for assistance with sampling, processing, and/or analysis. Zhaoying Li is thanked for her assistance in the graphic preparation. Williams S. Moore and Dalin Shi provided constructive comments on the manuscript. John Hodgkiss is thanked for his help with English. Comments from Wiley Evans and another anonymous reviewer have significantly improved the quality of the paper.

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