Research paper

Clay mineralogy indicates the Holocene monsoon climate in the Changjiang (Yangtze River) Catchment, China

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ABSTRACT

The sediments from two cores (CMS7 and LGZ) in the Changjiang (Yangtze River) Delta were collected for clay mineral analyses, with the aims of examining the origin of the clay and the application of clay mineralogy to the reconstruction of monsoon climate variability during the Holocene. The clay mineral assemblages of the Holocene sediments are similar overall to clays deposited in the modern Changjiang, albeit with large fluctuations in depositional environments. Peak contents of smectite occurring at ca. 13–11.5 ka are primarily due to the significant contribution of clays weathered from the upper Changjiang catchment under the impact of an enhanced Indian Summer Monsoon. The regular variations of crystallographic indices including illite crystallinity, chlorite crystallinity and chemical index of illite in the cores show close correspondence with the well-known oxygen isotopic curves of stalagmites in China. This suggests that the evolution of the Asian Summer Monsoon in the Changjiang catchment can be reliably reconstructed from variation in the weathering characteristics of the detrital clay component of sediments, despite the complex sediment source-to-sink transport patterns and the changes of depositional environments. For short timescales, the degree of crystallinity of clay minerals is more sensitive to chemical weathering and climate change than their absolute abundance. This study sheds new light on the reconstruction of paleoclimatic change in large drainage basins through the clayey sediments accumulated in estuarine and deltaic areas where land and sea interact.

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1. Introduction

Although weaker in amplitude than the large climate fluctuation during the last glacial cycle, the variability of climate during the Holocene is considered to be larger and more frequent than previously recognized (Alley and Ágústsdóttir, 2005; An et al., 2000; Bond et al., 2001; Denton and Karlén, 1973; Dykoski et al., 2005; Fleitmann et al., 2007; Hong et al., 2005; Kleinen et al., 2011; Mayewski et al., 2004; Wang et al., 2005). Changes in solar insolation linked to Earth’s orbital variations and solar intensity variability primarily account for global climate change during the Holocene (Dykoski et al., 2005; Hong et al., 2005; Mayewski et al., 2004; Wang et al., 2005). Climate change significantly affects biogeochemical evolution and human civilization in the Holocene (Chen et al., 2005b, 2008; deMenocal et al., 2000; Hodell et al., 1991), and thus, has attracted increasing research attention in recent decades. Meanwhile, various archives, such as stalagmite (Dong et al., 2010; Dykoski et al., 2005; Hong et al., 2005; Wang et al., 2005), lacustrine and estuarine sediments (Chen et al., 2005a; Shi et al., 1993; Xiao et al., 2006; Zhang et al., 2011; Zong et al., 2006), palynological data (Atahan et al., 2008; Li et al., 2010; Liu et al., 1992, 1998; Shen et al., 2006; Yi and Saito, 2004; Yi et al., 2003a,b, 2006), ice core (Shi et al., 1999; Thompson et al., 2006), and ancient literatures (Chu, 1973; Zhang, 2006), have been used to reconstruct the Holocene climate in China, with emphasis on the spatial and temporal variability of the East Asian Summer Monsoon (EASM). In Liu et al. (1992, 1998), high abundance of pollen and spore between 10,000 and 4800 years B.P. suggests that the EASM during the early-mid Holocene probably extended beyond its present boundary to reach western Tibet in response to orbital forcing. An et al. (2000) suggested that peak precipitation of the EASM in the Holocene was asynchronous in central and eastern China, reaching the maximum at different times in different regions. In Wang et al. (2005) and Dykoski et al. (2005), the EASM intensity broadly follows the summer insolation. Zong et al. (2006) suggested that organic carbon compositions and diatom abundances in the Holocene sediments of the Zhujiang (Pearl River) Estuary can indicate the evolution of the EASM. The Changjiang (Yangtze River) as the largest river originating from the eastern Tibetan Plateau, delivers a huge amount of terrigenous sediment from its vast catchment into the East China Sea, and has developed a large delta in the river mouth and unique
sedimentary systems on the shelf. The sediment that rapidly accumulated in the delta area during the Holocene is up to about 70 m in thickness (Hori et al., 2001a,b; Li et al., 2000), which makes the delta a natural laboratory for the study of land–sea interaction and paleoenvironmental change. Although the Holocene climate in China has been extensively studied as introduced above, only a few studies have paid special attention to the Changjiang catchment and delta area. This is mostly because of the lack of high quality research materials, reliable proxies, good age control of sedimentary strata and few high-resolution sampling analyses. In particular, sedimentary environments in the delta and estuary are more dynamic and interactive than those in the inland region and open ocean, and strong land–sea interaction results in frequent occurrence of hiatus. Previous research on the deltaic sediments used either a palynological approach or organic geochemical proxies to reconstruct the Holocene climate in the Changjiang catchment (Atahan et al., 2008; Chen et al., 2009; Liu et al., 1992, 1998; Yang et al., 2011; Yasuda et al., 2004; Yi and Saito, 2004; Yi et al., 2003a,b, 2006). From these studies, the drill core CM97 from Chongming Island in the Changjiang river mouth (Fig. 1) is now regarded as a classical borehole, and has been widely investigated in terms of its sediment stratigraphy and depositional facies (Hori et al., 2001a,b, 2002a,b), and environmental changes (Yang et al., 2011; Yi and Saito, 2004; Yi et al., 2003a,b, 2006). The core sediments are dated back to about 13 ka and allow us to plot the climate evolution through the Holocene in the Changjiang catchment.

In this paper, clay mineral assemblages of the sediments from Core CM97 were investigated and the proxies of clay mineral crystallinity were applied to link the weathering process, sediment transport and Holocene monsoon climate in the Changjiang drainage basin.

2. River setting

The Changjiang River originates from the northeast of the Tibet Plateau with an elevation of 6621 m and a drainage area of about 1.8 × 10^6 km^2, and flows about 6390 km eastward to the East China Sea (Fig. 1). The river delivers about 390 × 10^6 ton/year of suspended sediment and 896.4 × 10^9 m^3/year of water into the estuary based on the hydrological observation from 1950 to 2010 at Datong Gauge Station (China Ministry of Water Resources, 2011).

Geologically, the Changjiang catchment is primarily situated on the Yangtze Craton, and the upper basin was greatly affected by uplift of the Tibet Plateau during the Cenozoic, and the mid-lower basin was mostly shaped by the Paleozoic and Mesozoic tectonic movements. Diverse source rock types characterize the Changjiang catchment, which predominantly comprise Archean metamorphic rocks, Paleozoic carbonate and siliciclastic rocks, Mesozoic and Cenozoic igneous and clastic rocks, and Quaternary detrital sediments.

The Changjiang catchment is strongly influenced by a subtropical monsoon climate, which causes large seasonal and spatial variability in temperature, precipitation, runoff and fluvial discharge. The EASM played a key role in the climate evolution of the Changjiang basin during the Holocene, while the upper catchment may have been affected by the Indian Summer Monsoon (An et al., 2000; Dykoski et al., 2005; Mao et al., 2010).

3. Sample sources and analytic methods

Two cores were collected from the Changjiang Delta for the study of clay mineralogy. Core CM97 was drilled on Chongming Island in

![A schematic map of the Changjiang Delta with the locations of Core CM97 and Core LGZ.](image)
the present river mouth and the core length is 70.0 m. A short core, LGZ, of 2.0 m length was taken in a newly-formed bar in the lower Changjiang mainstream, about 150 km upstream from the river mouth (Fig. 1). Core LGZ is composed predominantly of clayey silt which accumulated over the last 150 years in the lower Changjiang mainstream, based on the 210Pb geochronology (Zhan et al., 2010).

The sedimentary sequences of Core CM97 primarily consist of fluvial or river channel facies at the base (70.0–603.3 m in burial depth), with floodplain facies (60.3–43.2 m), estuarine facies (43.2–30.0 m), neritic facies (30.0–20.1 m) and deltaic facies at the top (20.1–0.0 m) (Fig. 2). Detailed sediment stratigraphy, delta evolution and sedimentary environment changes in the Changjiang delta and estuarine area during the postglacial period have been reported previously (Chen et al., 1999). Similarly, the ChC was measured on the 7 Å peak (Árkai and Ghabrial, 1997; Árkai et al., 1995; Mas et al., 2006) and also expressed in °Δ2θ. The CII refers to the ratio of the 5 Å and 10 Å peak areas. Ratios for the CII <0.5 may represent Fe–Mg-rich illite, while ratios >0.5 are primarily found in Al-rich illite (Mao et al., 2010).

4. Results

The sediment grain size and clay mineralogical composition of Core CM97 are shown in Fig. 2. The mean grain size (Mz) varied between 2.1 Φ and 7.3 Φ and exhibited irregular downcore variation. The bottom fluvial facies consisted of the coarsest sandy sediments with minor gravels, with a mean size of 4.5 Φ. The mean grain size gradually became finer from the fluvial facies upward to the neritic facies and the finest sediments were about 7.0 Φ in Mz. The deltaic facies at the top, however, became coarser and the average Mz was about 5.1 Φ. As shown in previous studies on clay mineralogy of the Changjiang-derived sediments (Dou et al., 2010; Fan et al., 2001; Gao et al., 2004; Mao et al., 2010; Wu et al., 2011; Yang et al., 2003), the clay minerals of Cores CM97 and LGZ sediments were dominated by illite, ranging from 19.8% to 68.7% and averaging at 56.7%. Kaolinite and chlorite yielded similar contents in the core sediments, with averages of 18.4±2.9% and 15.2±3.0% respectively. In comparison, smectite was less abundant but exhibited the largest downcore variation (2.1–70.0%) in Core CM97, and especially, reached the highest average content of 28.6% in the fluvial facies (Fig. 2).

In Core CM97, the illite crystallinity (IC) varied between 0.26 and 0.51 (°Δ2θ) and chlorite crystallinity (ChC) between 0.29 and 0.58 (°Δ2θ), while the chemical index of illite (CII) ranged from 0.32 to

![Fig. 2. Lithology of Core CM97 and depth profiles of mean grain size (Mz) and clay mineral contents. The lithology was modified from Hori et al. (2001b) and the age model (in calendar years before present) from Yi et al. (2003b).](image-url)
1.04. Based on the downcore variations of clay mineral content and crystallinity index, Core CM97 was subdivided into four units: Unit 1 (0–16 m), Unit 2 (16–42 m), Unit 3 (42–60 m) and Unit 4 (60–70 m) (Fig. 3). The indices of IC, ChC and CII showed large but overall similar downcore fluctuations, with higher values in Unit 2 and lower in Unit 4. Remarkably, increasing trends of IC, ChC and CII values were clearly observed from Unit 4 to the lower part of Unit 3 and also from the upper Unit 3 to the middle part of Unit 2, whereas the obvious decrease of these indices occurred at the transition from Unit 2 to Unit 1.

5. Discussion
5.1. Provenance of clay minerals in the Changjiang Delta during the Holocene

Clay mineral assemblages in coastal and marine sediments are predominantly determined by sources of clayey sediment, provenance weathering, sediment transport and depositional processes (Chamley, 1989; Gao et al., 2004; Pandarinath, 2009; Pandarinath et al., 1999). Estuarine and delta areas of large rivers receive huge detrital loads from their vast drainage basins that can encompass a variety of geological, geographical and climatic environments. Consequently, the clayey sediments accumulated in the estuarine and delta areas typically represent an average composition of clay minerals sourced from the whole catchment (Singer, 1980, 1984).

The clay mineral assemblages of Cores CM97 and LGZ are overall similar to Changjiang-derived clays reported in previous studies (Fig. 4) (Dou et al., 2010; Fan et al., 2001; Fang et al., 2007; Gao et al., 2004; Mao et al., 2010; Wu et al., 2011; Yang, 1988; Yang et al., 2003). In the ternary diagram of (illite + smectite)–chlorite–kaolinite (Fig. 4), most of the samples in Core CM97 plot in the compositional range of Core LGZ (dashed circle) albeit with relatively scattered distribution. The sediments in Core LGZ that were deposited in the lower Changjiang mainstream over the last 150 years (Zhan et al., 2010), are definitely derived from the large drainage basin. Therefore, it is inferred that the Holocene clays accumulated in the Changjiang Delta have similar composition on average to the present-day clay minerals in the Changjiang sediments. In Yang et al. (2001), geochemical data also indicate that the fine-grained sediments (<0.063 mm) of Core CM97 have similar composition to the modern fluvial sediments, diagnostic of their genetic inheritance.

Nevertheless, the four clay minerals in Core CM97 all show similar large variation in their percentages in the floodplain and fluvial facies (Units 3 and 4; Table 1; Fig. 2). The core sediments were deposited in various environments, from terrestrial (floodplain and fluvial facies), coastal (deltaic and estuarine facies) to shallow marine (neritic facies). Different hydrodynamic forces in these depositional environments may complicate the distribution of clay minerals, although they were all sourced from the Changjiang catchment. The clay mineral assemblages in the deltaic, neritic and estuarine facies (Units 1 and 2) are relatively stable and largely overlap with those from Core LGZ sediments. In comparison, the clays in the terrestrial floodplain and fluvial facies (Units 3 and 4) have different mineralogical characteristics especially in the relative amounts of illite and smectite (Fig. 4). Different hydrodynamic sorting and/or diagenesis in these variable depositional environments may account for this difference. In general, estuarine and neritic and part of deltaic environments are characterized by relatively deeper waters than those in floodplain and fluvial environments. The multi-sourced clayey sediments derived from the Changjiang catchment might have been well mixed in the marginal marine environments whereas the clays in the floodplain and river channel may better reflect their source characteristics due to rapid transport and deposition. Influence of hydrodynamic sorting on clay minerals in coastal and marine environments has been suggested by previous workers (Chamley, 1989; Dou et al., 2010; Gao et al., 2004; Steinke et al., 2008). In particular, smectite which has the smallest size of all clay minerals, is prone to winnowing by oceanic currents (Chamley, 1989). Nevertheless, the large fluctuation and extraordinary high content of smectite in floodplain and fluvial facies are primarily due to the changing provenances of clayey sediments in the late deglaciation (ca. 13–11 ka) rather than winnowing process during sediment transport and deposition. Sedimentary smectite is mostly derived from temperate–humid or from arid weathering processes, or from mafic and older sedimentary rocks (Chamley, 1989; Thiry, 2000). The Emeishan basalt in the upper Changjiang valley is regarded as the only large igneous province in China and one of the largest basalt provinces in the world (Xiao et al., 2003). The weathering of basalts may provide plenty of smectite for the upper Changjiang River, which has been evidenced from higher smectite contents in the upper Changjiang sediments (Mao et al., 2010; Wu et al., 2011). An et al. (2000) suggested that the Holocene climate optimum, as defined by peak summer monsoon precipitation, was asynchronous in central and eastern China, reaching a maximum at different times in different regions. In southwestern China the maximum

![Fig. 3. Downcore variations of illite crystallinity, chlorite crystallinity and chemical index of illite in Core CM97.](image-url)
precipitation appeared ca. 11,000 years ago, which was probably related to the maximum landward extension of the Indian Summer Monsoon. Therefore, the intensified Indian Summer Monsoon during the late postglacial period to early Holocene (ca. 13–11 ka) might have brought a large amount of precipitation to the upper Changjiang valley, which promoted the weathering of Emeishan basalt and other basic rocks to smectite. This may explain the peak amounts of smectite in the floodplain and fluviatile facies (Units 3 and 4) of Core CM97, while other samples in these two units have low smectite abundance, indicative of the complicated sediment source-to-sink transport pattern in the large Changjiang catchment.

5.2. Clay mineralogy indicating the monsoon evolution during the Holocene

Clay minerals in sediments can provide important constraints on continental weathering under different climate regimes (Biscaye, 1965; Chamley, 1989; Petschick et al., 1996; Singer, 1980, 1984; Thiry, 2000). Recent studies suggest that clay mineralogy in Asian marginal seas can indicate the evolution of EASM on orbital scale (Boulay et al., 2005; Liu et al., 2003, 2005; Wan et al., 2010), but primarily reflects sediment sources and hydrodynamic fractionation over shorter time scales such as the last glacial-interglacial interval (Dou et al., 2010; Steinke et al., 2008).

Schematically, illite and chlorite are interpreted to form in relatively very cold or hot-dry climate, while a hot and humid climate leads to stronger chemical weathering and the formation of kaolinite. Smectite minerals consist of several subgroups which may form in different chemical and climatic environments. Temperate climate tends to form incompletely altered minerals, mostly exfoliated illite and chlorite, irregular mixed-layers, vermiculite and degraded smectite, depending on the local climate. Sub-arid areas give way to well-crystallized Fe-smectite (Chamley, 1989; Singer, 1980, 1984). In fact, clay mineral assemblages in soils and sediments may not directly indicate chemical environments and paleoclimate but integrate records of different climatic impacts. Differentiation of clay assemblages during sediment transport and deposition, due to differential flocculation or size sorting, may significantly alter climate-induced signals, especially for the clays derived from large drainage basins and from multi sources. Therefore, crystallographic indices of clay minerals such as illite crystallinity (IC), chlorite crystallinity (ChC) and chemical index of illite (CII) are less influenced by sediment dynamic differentiation, and can better record climate signals than the absolute amounts (clay assemblages). These indices, as the characterization of mineral crystallinity degree, can reflect weathering conditions of sediment source area even if clay mineral contents are changed by hydrodynamic sorting during transport and deposition. In general, lower values of IC and ChC represent higher crystallinity, characteristic of weak hydrolysis in continental sources and dry and cold climate, while higher IC and ChC values indicate lower crystallinity, standing for strong hydrolysis caused by humid and warm climate (Chamley, 1989; Pandarinath, 2009; Pandarinath et al., 1999).

In the present day, the large Changjiang catchment covers different climate regimes with wide variation in temperature, precipitation and vegetation type. The upper part runs mainly through the Qinghai-Tibet Plateau and Yunnan–Guizhou Plateau, where the exposed rocks easily produce clays by strong physical erosion and limited chemical weathering under rigid climate. In contrast, the middle–lower basin is subject to subtropic climate with much higher annual precipitation and higher temperature on average than in the upper basin, which accounts for relatively strong chemical weathering and hydrolysis. Consequently, clay mineralogical indices of IC, ChC and CII yield lower values or higher crystallinity in the sediments of the upper Changjiang reaches, and higher values or poor crystallinity in the middle–lower reaches (Mao et al., 2010).

The crystallographic indices of IC, ChC and CII all exhibit similar and regular variations in Core CM97 (Fig. 3), showing higher values in Unit 2 (dominated by neritic and estuarine facies) and the lowest in Unit 4 (fluviatile facies). Of particular note is the correspondence of the downcore variations of IC, ChC and CII with the well-known oxygen isotope curves of Dongge stalagmite in Guizhou Province, Southwest China (Dykoski et al., 2005; Wang et al., 2005) and of Sanbao Cave in Hubei Province, central South China (Dong et al., 2010) (Fig. 5). The oxygen isotopic records of these Chinese stalagmites are regarded as the best indication of Asian summer monsoon precipitation during the Holocene. Correspondingly, we infer that the indices of IC, ChC and CII in Core CM97 can also indicate the evolution of Holocene monsoon climate within the Changjiang catchment.

During the late deglaciation (about 13–11.5 ka, Units 3 and 4), the climate was colder and drier than at present; a weakened EASM (Dong et al., 2010; Dykoski et al., 2005) led to reduced precipitation in the Changjiang catchment. The sediments weathered in this climate have low IC, ChC and CII values (Fig. 5). The pollen data of Core CM97 also suggests cold and dry climate at ca. 13–11 ka (Yi et al., 2003b). The ratio of total organic carbon to total nitrogen (TOC/TN) exhibits extraordinary high values in Unit 4 (fluviatile facies), suggesting the proximal input of terrestrial organic matter (Yang et al., 2011).

With the onset of Holocene, the enhanced EASM arrived at the maximum at about 9 ka and then gradually weakened towards the late

Fig. 4. Ternary diagram showing the comparison of clay mineral assemblages between the sediments from Core CM97 (shown as different depositional facies), Core LGZ (marked with dashed ellipse) and modern Changjiang River (Dou et al., 2010; Yang et al., 2003). K: kaolinite; C: chlorite; S: smectite; I: illite.
Table 1  
Relative proportions of clay minerals in the <2 μm size fraction of the sediments from Cores LGZ and CM97 and crystallinity indices.

<table>
<thead>
<tr>
<th>Cores</th>
<th>Depth (m)</th>
<th>Illite (%)</th>
<th>Kaolinite (%)</th>
<th>Chlorite (%)</th>
<th>Smectite (%)</th>
<th>IC (ΔΔm)</th>
<th>ChCl</th>
<th>CII</th>
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<tbody>
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Note: IC = Illite Crystallinity; ChCl = Chlorite Crystallinity; CI = Chemical Index of Illite.

Holocene (An et al., 2000; Dong et al., 2010; Dykoski et al., 2005; Wang et al., 2005). The strengthening EASM climate might have caused abundant rainfall and enhanced chemical weathering in the Changjiang catchment, especially in the middle-lower basins, which consequently resulted in a significant supply of poorly-crystallized clay minerals.
with high IC, ChC and CII values into the Changjiang river mouth (Unit 2, Fig. 5). Pollen and organic geochemical data further imply that the paleoclimate was warm and humid at ca. 5–4 ka (Yi et al., 2003b), and marine organic matter dominated the estuarine to shallow marine depositional environments (Yang et al., 2011).

Over the past 1500 to 2000 years, the EASM was weaker than in the early-mid Holocene, and the Changjiang Delta rapidly prograded with the transition from shallow marine to deltaic sedimentary environment (Hori et al., 2001a,b, 2002a,b; Li et al., 2000; Yang et al., 2011). The deltaic sediments (Unit 1) have lower IC, ChC and CII values relative to the shallow marine and estuarine sediments (Unit 2) (Fig. 5), also suggesting weaker chemical weathering with the decrease of monsoon precipitation during the late Holocene. Another possibility regarding the origin of clays in Unit 1 is that increasing anthropogenic activity over the last 2000 years caused strong erosion in the Changjiang catchment, especially in the upper basin (Saito et al., 2001), which resulted in the dominance of the upper Changjiang-derived sediments in the deltaic deposition since 2 ka. The clay minerals derived from the upper Changjiang reaches generally have low IC, ChC and CII values because of weak chemical weathering under an unfavorable climate regime. The crystallographic indices including IC, ChC and CII of clay minerals from the Holocene sediments in the Changjiang Delta appear to closely reflect changes in the paleoclimate across the large drainage basin, although the resolution of change is less precise than that obtained from the well-known stalagmite records.

**6. Conclusions**

The clay mineral compositions of two cores from the Changjiang Delta were investigated in order to examine the origins of the clays deposited in the Changjiang estuary and delta, and to evaluate the impact of monsoon climate on the weathering, transport and deposition of clays in the catchment during the Holocene. The major clay mineral assemblages of the Holocene sediments are overall similar to the clays of the modern Changjiang, dominated by illite, but exhibit large fluctuations in terrestrial depositional environmental (fluvial to floodplain facies) in the early Holocene. The estuarine, shallow marine and deltaic environments are characterized by relatively stable clay mineralogy. Independent of the absolute abundance of clay minerals, the crystallographic indices of illite crystallinity, chlorite crystallinity and chemical index of illite, show regular variations in Core CM97 that were used to identify four depositional units with unique clay mineral patterns.

The crystallographic indices of illite and chlorite in sediments as the indication of clay mineral crystallinity, are closely related to the continent weathering and climatic environments. The variations of these indices in the Holocene sediments in the Changjiang Delta are closely comparable with the well-known oxygen isotopic curves of cave carbonates in southeast China. The results suggest that the crystallographic indices of specific clay minerals can distinctly indicate monsoon climate variability in the large drainage basin. Pollen and organic data of the core sediments further indicate similar paleoclimatic changes in the catchment and delta during the Holocene. In particular, weakening in late deglaciation (ca. 13–11.5 ka) and strengthening in the early-mid Holocene (ca. 11.5–4 ka) of EASM would be expected to have resulted in a large variability in precipitation and temperature over the vast catchment, with a consequent change in the intensity of chemical weathering, as evidenced by the fluctuation of crystallographic indices of the clay minerals.

The clay mineral assemblages are primarily determined by the various sources of clays in the Changjiang catchment and transport and depositional processes, and thus, cannot distinctly reflect EASM evolution in the Holocene. Smectite, for example, has the largest variation in concentration of the clays deposited from 13,000 to 11,300 cal years B.P., probably reflecting the significant contribution of clays from the upper Changjiang valley with the enhancing Indian Summer Monsoon at that time.

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