## 石英の ESR 信号強度を利用した砕屑物の供給源推定と東アジアの古気候復元

Estimation of detrital material provenance based on ESR signal intensity and reconstruction of paleoclimate in East Asia

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#### Abstract

East Asian Summer Monsoon (EASM) front penetrates deeper into inland China with an increase in its intensity. Such changes in EASM intensity influence spatial pattern of summer precipitation in East Asia. Since EASM brings most of annual precipitation in East Asia, it is important to understand spatio-temporal changes in EASM precipitation. The Yangtze River, which occupy a large part of South China, discharges large amount of sediments to East China Sea and the provenance of sediments reflects the precipitation patterns within the drainage. In this study, the spatial variation of EASM during the last 6 ky are reconstructed based on the change in the provenance of sediment in the cores recovered from the Mud Belt of the East China Sea shelf.

Keyword: East Asian Summer Monsoon, Electron spin resonance, quartz

#### 1. Background

Hydroclimate variations associated with the East Asian Summer Monsoon (EASM) precipitation exert significant impacts on lives of over 450 million people inhabiting within the Yangtze River drainage. Observational data clearly demonstrate that seasonal shift of main precipitation area is attributable to the reposition of northern limit of summer monsoon, which would have lead to provenance and composition changes of suspended particulate materials (SPMs) transported by the Yangtze River. Therefore, the decadal to millennial time-scale variability of the position of the EASM rain belt mentioned above could be recorded as the change in compositional variation of sediment originated from the SPMs in the Yangtze River. The subaqueous Yangtze delta and East China Sea (ECS) shelf are primary depositional sites owing to massive inputs of terrestrial materials from the Yangtze River. We expect to recognize provenance changes of SPMs in association with EASM precipitation variability by examining the sediments of MD06-3040 core taken from the Mud Belt of the inner shelf of ECS on the basis of the electron spin resonance (ESR) signal intensity and crystallinity index (CI) of fine silt fraction of quartz, which should represent SPMs discharged from the Yangtze River.

### 2. Sample and Methods

MD06-3040 core was recovered from the inner shelf of East China Sea by Marion du Fresne cruise in the summer of 2006. Its length is 19.36m and its water depth is 47m. Eighty eight bulk samples from core MD06-3040 covering last 6 kyr was selected for provenance analysis of quartz grains. Pretreatment was conducted to remove organic matter, iron and manganese oxides and hydrooxides, and carbonate minerals. After pretreatment, samples were separated into 4-16  $\mu$ m and 16-63  $\mu$ m size fractions using the syringe and pipette methods. ESR and CI were determined for these two grain size fractions. The ESR signal intensity of the E'1 center of quartz, an unpaired electron in a single silicon sp3 orbit oriented along a bond direction into an oxygen vacancy <sup>[1]</sup>, was used to estimate the relative number of oxygen vacancies in quartz. Oxygen vacancies in quartz have been formed by natural radiation, and are known to increase with increasing age of the host rock <sup>[2,3]</sup>. Consequently, higher values of ESR signal intensity of the E'<sub>1</sub> center in quartz mean the older ages of the host rocks.

Firstly, pretreated samples were irradiated with  $\gamma$ -radiation (total dose of 2.5 kGy) using a <sup>60</sup>Co source at the Inter-University Laboratory for the Joint Use of Japan Energy Agency Facilities. Subsequently, Atomic pretreated samples were heated at 300 °C for 15 minutes to convert the oxygen vacancies to E'<sub>1</sub> centers <sup>[4]</sup>. ESR signal intensity measurements were conducted at room temperature with an X-band ESR spectrometer JEOL JES-FA100 at University of Tokyo under 0.01 mW of microwave power, 0.1 mT magnetic field modulation (100 kHz), 5 mT scan range, two minutes scan time, and 0.03 seconds time constant. ESR intensity of the E'1 centers was normalized to the quartz content of each sample to estimate the ESR signal intensity of pure quartz. The ESR signal intensity of quartz is expressed in spin units: one spin unit is equivalent to  $1.3 \times 10^{15}$ spins/g<sup>[5]</sup>. The reproducibility of ESR signal intensity was smaller than  $\pm 1.5$  spin units.

### 3. Results and Discussions

ESR signal intensity and CI of quartz in the sediments discharged from the major tributaries of the Yangtze River were already examined and the result was reported

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by us <sup>[6, 7]</sup>. In general, the western and northeastern tributaries yield the sediment with lower (0~10) ESR signal intensity, while the southeastern tributaries tend to yield the sediments with higher (>10) ESR signal intensity (Fig. 1). When the increase in EASM intensity causes the increase in precipitation in the western and northern parts of Yangtze River basin, it is expected that the western and northeastern tributaries yield more sediments and that ESR signal intensity of sediments discharged from the Yangtze River mouth becomes lower. On the contrary, it is expected that ESR signal intensity of the sediments discharged from the Yangtze River mouth becomes higher when EASM becomes weak and the precipitation in the southeastern part of the Yangtze River basin increases.

In order to discriminate the sediment contributions from different parts of Yangtze River basin, we compared the ESR signal intensity and CI of quartz in the fine silt fraction of the sediments from core MD06-3040 with those of the modern Yangtze River sediment samples.

The ESR values of quartz in fine silt fraction of core MD06-3040 samples showed larger values compared to the ESR value in coarse silt fraction during 6.5 to 6 cal kyr BP, 6 to 4 cal kyr BP, and 1.8 to 1 cal kyr BP. The ESR values in both fine silt fraction and coarse silt fraction of core MD06-3040 samples have similar values during 4 to 1.8 cal kyr BP and 1 to 0 cal kyr BP. Moreover, detailed examination of quartz provenance within the Yangtze River drainage using ESR and CI enable us to discriminate the sediment contributions from the uppermost-upper reaches (northeastern and western tributaries) versus middle-lower reaches (southeastern tributaries) of the Yangtze drainage (Fig. 1).

This observation suggested that variability of the major location of EASM precipitation (EASM rain front) on multi-centennial to millennial-scale has been detected from ESR vs CI plot in Figure 1, which showed SPM provenance from the southeastern tributaries of Yangtze with some contribution from western and northeastern tributaries from 6 to 4 cal kyr BP, dominantly from the western and northeastern tributaries from 4 to 1.8 cal kyr BP, mixed contribution between western- northeastern tributaries and southeastern tributary from 1.8 to 1.0 cal kyr BP, and mixed contribution between westernnortheastern tributaries and southeastern tributary with slightly increased contribution from southeastern tributaries from 1.0 to 0.6 cal kyr BP, respectively. This in turn suggests more precipitation in the southeast area of Yangtze River drainage from 6 to 4 ka, in the western to northwestern area from 4 to 1.8 ka, and in wider area between the northeastern and southeastern during 1.8~0.6 ka with increased contribution from southeastern area after 1.0 ka. The result further suggests the deepest penetration of EASM front into China during 4~1.8 ka.



Figure 1. ESR signal intensity and Crystallinity Index of quartz in the fine silt fraction of the sediments from MD06-3040 core during last 6 ky compared with those in the fine silt fraction of the river bed sediments from

Yangtze River Basin<sup>[7]</sup>.

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