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Significant changes in land vegetation and oceanic redox across the Cretaceous/Paleogene boundary

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ABSTRACT

The impact of an extraterrestrial body 65.5 Ma caused the so-called Cretaceous/Paleogene (K/Pg) event that resulted in a mass extinction and rapid changes in the surface environment on Earth. Here we report changes in land vegetation and oceanic redox across the K/Pg boundary at Caravaca, southeastern Spain, using biomarkers. The results reveal that the basal 3-mm-thick layer of the K/Pg boundary clay is marked by a rapid increase in the concentration of terrestrial long-chain *n*-alkanes and dibenzofuran, indicating the destruction of land vegetation and an increase in the supply of terrestrial organic matter into the marine environment during the deposition of the K/Pg boundary clay. This layer also records a rapid increase in the concentration of dibenzothiophenes, which indicates a change in redox conditions from oxic to anoxic/euxinic conditions in the intermediate water over the seafloor. The low-oxygen conditions could have been caused by an increase in the influx of terrestrial organic matter into the ocean. A rapid increase in the concentration of retene and retene/cadalene occurred during the deposition of the upper part of the boundary clay 10 kyr after the mass extinction, indicating the recovery of conifers, which greatly decreased at the K/Pg boundary.

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1. Introduction

The end-Cretaceous mass extinction is one of the five largest mass extinctions in Earth's history (Raup and Sepkoski, 1982). Previous research has shown that more than 60% of Cretaceous species disappeared at the K/Pg boundary (Schulte et al., 2010). It is widely accepted that the impact of an extraterrestrial body was a major factor in the mass extinction event (e.g., Alvarez et al., 1980). Schulte et al. (2010) examined global stratigraphic records across the K/Pg boundary and concluded that the Chicxulub impact triggered the K/Pg mass extinction. According to paleoenvironmental studies, the impact resulted in various environmental changes, such as global fires (e.g., Wolbach et al., 1985), global dust cloud (darkness and cooling), sulfate aerosols (darkness and cooling), and acid rain (e.g., Sigurdsson et al., 1992; Pope et al., 1994; Hildebrand, 2007). As a result of these environmental changes, land vegetation collapsed at the K/Pg boundary (e.g., Wolfe and Upchurch, 1986). In North America, palynological and megafloral paleobotanical studies have demonstrated vegetation changes coincident with the iridium anomaly. However, apart from the North American record, there are few reports of floral change across the K/Pg boundary (Nichols and Johnson, 2008). More studies of plants are necessary to elucidate global vegetation changes across the K/Pg boundary.

Ecological collapse and environmental changes in the ocean have also been reported (e.g., Kaiho et al., 1999). Darkness caused by the impact stopped photosynthesis, thus breaking down the food chain and causing the extinction of many marine organisms. In the Caravaca section in southeastern Spain, the abrupt extinction of planktonic foraminifera and calcareous nanoflora coinciding with the impact has been reported (e.g., Kaiho and Lamolda, 1999; Lamolda et al., 2005). Although the nanoplankton extinction was a global event, its intensity differed in various regions. Jiang et al. (2010) reported that nanoplankton extinction rates in the northern hemisphere were higher than those in the southern hemisphere. Dissolved oxygen in intermediate water changed across the K/Pg boundary in the ocean. Low oxygen conditions in the earliest Paleogene intermediate water were indicated by sulfur isotope analysis of whole rock sulfide in the Kawaruppu section (Kajiwara and Kaiho, 1992). Kaiho et al. (1999) also reported that intermediate water oxygen minima were widely developed during the earliest Danian, as evidenced by the Benthic Foraminiferal Oxygen Index, total organic carbon (TOC), the ratio of thick-walled oxic foraminifera relative to all oxic foraminifera, the hydrogen index, the oxygen index, Mn content, and $\Delta\delta^{34}S_{\text{sulfide}-\text{sulfate}}$ from Caravaca, Spain. However, whether intermediate water euxinia was a global event is still debated.

Some organic geochemical studies have sought to elucidate the environmental changes caused by the impact of an extraterrestrial body. Arinobu et al. (1999) identified a polycyclic aromatic hydrocarbon (PAH) peak that was evidence of forest fires at the K/Pg boundary. Sepúlveda et al.'s (2009) analysis of the stable isotopes of carbon and

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nitrogen and the abundances of algal steranes and bacterial hopanes indicated the quick recovery of algal primary productivity less than a century after the K/Pg boundary. Furthermore, Yamamoto et al. (2010) reported plant diversification processes in the early Paleogene based on leaf wax *n*-alkanes. Such biomarker analyses can help with paleoenvironmental reconstructions. However, organic geochemical studies of the K/Pg boundary remain inadequate.

In this report, we provide the first high-resolution profiles of the changes in the distribution of biomarkers, indicating changes in land vegetation and oceanic redox across the K/Pg boundary at Caravaca, which represents one of the most complete and least disturbed K/Pg sections in the world (Canudo et al., 1991; MacLeod and Keller, 1991).

2. Sampling

The Caravaca K/Pg section (Fig. 1) is located in the Betic Cordillera of southeastern Spain (38°04′35″N, 1°52′40″W). In the K/Pg section at Caravaca, marlstones of Cretaceous age are lithologically separated from marlstones of Paleogene age by a 7–10 cm thick, dark, clay–marl bed (the boundary clay layer). Within the boundary clay layer, a 1–2 mm, rust-orange, basal layer referred to as the red layer (or fallout lamina) contains the iridium (Ir) anomaly (Smit and Ten Kate, 1982) and is underlain by a 3-mm greenish transition layer. In this paper, the base of the red layer is defined as the K/Pg boundary; set to 0 cm, with the heights of the lower and upper strata being described relative to it. The Caravaca section represents paleowater depths of 200–1000 m (Coccioni and Galeotti, 1994; MacLeod and Keller, 1994).

We collected the sedimentary rock samples for biomarker analysis after removing the surface of the outcrop to 40–50 cm depth. Weathering is shallow because soft marly rocks are constantly removed from the small creek where the samples were taken. This is typical of the badlands in this Mediterranean climatic region, with no vegetal cover on the studied exposures.

3. Methods

3.1. Extraction and separation

For organic geochemical study, 28 clay or marl samples were collected from the 37-cm-thick sedimentary sequence (Figs. 2, 3). Approximately 50, 25, or 15-g hand-sized specimens were powdered for each sample after removing apparent surface contamination. The powdered samples were extracted for 48 h using a Soxhlet apparatus and a dichloromethane:methanol mixture (7.5:1 v/v). The extracts were dried over Na_2SO_4 and concentrated by evaporation under reduced pressure. The concentrated extracts were separated into nine fractions on a silica gel column (0.6 g of silica, 63–200 mesh) by elution with the following solvents: 2 ml of *n*-hexane (F1a), 4 ml of *n*-hexane (F1b), 3 ml of *n*-hexane/toluene 3:1 v/v (F2), 3 ml of *n*-hexane/ethyl acetate 19:1 v/v (F3), 3 ml of *n*-hexane/ethyl acetate 9:1 v/v (F4), 3 ml of *n*-hexane/ethyl acetate 17:3 v/v (F5), 3 ml of *n*-hexane/ethyl acetate 4:1 v/v (F6), 3 ml of *n*-hexane/ethyl acetate 3:1 v/v (F7), and 10 ml of methanol (F8). The aliphatic hydrocarbon fraction (F1a) and aromatic hydrocarbon fraction (combinations of F1b and F2) from each extract were analyzed by gas chromatography–mass spectrometry (GC–MS).

3.2. Analysis of organic compounds

Identification of the organic compounds was performed using an Agilent 6893 gas chromatograph interfaced to an Agilent 5973 mass selective detector (MSD), operated with an ionizing-electron energy of 70 eV and scanned from m/z 50 to 550 with a scan time of 0.34 s. A fused silica HP-5MS capillary column (30 m, 0.25 mm i.d., 0.25 μm film thickness) was used with helium as the carrier gas. Samples were injected at 50 °C and held at that temperature for 1 min; the temperature was then raised to 120 °C at a rate of 30 °C/min, then to 310 °C at a rate of 5 °C/min and finally held constant for 20 min.

3.3. Total organic carbon (TOC) measurement

The TOC content of the sediment was determined using a Carlo-Erba elemental analyzer (EA1108). The powdered samples (3 g) were decarbonated in 6 N HCl at 70 °C for 24 h. Acid-treated samples were washed in ultrapure water a few times to remove the acid. Approximately 10 mg of dried residual powder free of carbonate was packed into a tin capsule and introduced to the EA1108. Corrections of the carbon content were made for the weight loss incurred during acid treatment. The limits of detection for this analytical method are approximately 0.01 wt.% for carbon. The reproducibility of the carbon concentrations is better than 0.01 wt.%.

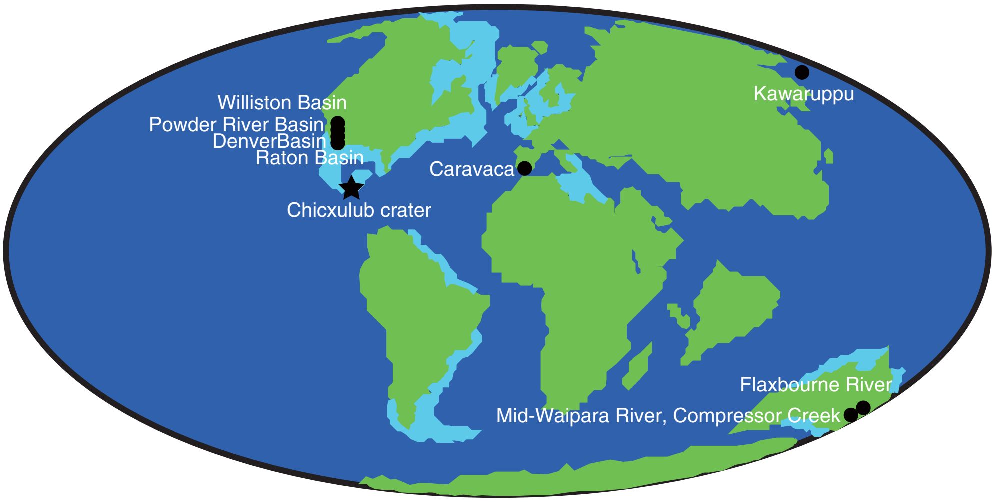

Fig. 1. Paleomap showing the studied section (Caravaca) and locations referred to in this paper.

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