

Comparison of elemental concentration in near-surface late Holocene sediments and precipitation regimes of the Yucatán Peninsula (Mexico): a preliminary study

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ABSTRACT

Paleoclimate research in the Yucatán Peninsula of Mexico has mainly involved fossil pollen preserved in sedimentary archives whilst geochemistry has received limited attention. In this study, we compared concentrations of K, Ti, Fe, Ca and Sr in 35 near-surface late Holocene sediments collected from different permanent and seasonal water bodies across the peninsula with mean annual precipitation of 600-1600 mm in order to explore the relationship between concentrations of inorganic elements and precipitation. The calcareous provenance has a diluting effect and sediments have less K, Ti and Fe (associated with clastics) and more Ca and Sr (associated with carbonate and evaporite) compared to the Upper Continental Crust. Individual elements and ratios of K/Ca, Ti/Ca and Fe/Ca in samples with minimal clastics did not show any relationship with precipitation. Average values of elemental ratios in sediments with more clastics increase as the mean annual precipitation increases. However, the sampling protocol and presence of most of the sampled water bodies being in a single precipitation cluster (i.e. 1200-1400 mm/year) did not allow a complete evaluation of the potential correlation between precipitation and elemental ratios. Considering that the concentration of Ti is low in sediments of the limestone-rich Yucatán Peninsula and Fe is mobile in anoxic depositional environments, the K/Ca ratio can be used as a preliminary proxy to evaluate precipitation in sedimentary records.

Keywords: geochemistry, element ratio, paleo-precipitation proxy, calcareous provenance, Yucatán Peninsula.

Comparación de concentración elemental en sedimentos superficiales del Holoceno tardío y los regímenes de precipitación en la Península de Yucatán, México: un estudio preliminar

RESUMEN

La investigación paleoclimática en la Península de Yucatán (YP) de México involucra principalmente el polen fósil preservado en archivos sedimentarios, mientras que la geoquímica ha recibido menos atención. En este estudio, se compararon las concentraciones de K, Ti, Fe, Ca y Sr, en 35 muestras superficiales de sedimento del Holoceno tardío, colectadas en un gradiente de precipitación entre 600-1600 mm/a, a lo largo de la YP, con el objetivo de establecer un testigo de la paleo-precipitación para la región. El origen calcáreo tiene un efecto de dilución en las concentraciones elementales, provocando que los sedimentos presenten baja cantidad de K, Ti y Fe (elementos asociados con clásticos) y mayor cantidad de Ca y Sr (elementos asociados con carbonato y evaporitas), en comparación con la corteza continental superior. Los elementos individuales y las relaciones de K/Ca, Ti/Ca y Fe/Ca en muestras con cantidades mínimas de clástico, no muestran una relación con la precipitación. Los valores promedio de la relación elemental en sedimentos con más clásticos, muestra tendencias positivas con la precipitación. No obstante, el protocolo de muestreo y la presencia de un

mayor número de cuerpos de agua investigados en un simple aglomerado (p. ej. 1200-1400 mm/a) no permitió completar la evaluación de la correlación potencial entre precipitación y la relación elemental. Considerando que la concentración de Ti en los sedimentos ricos en calizas de la PY es bajo y el Fe es móvil en ambientes de depositación anóxicos, la relación K/Ca puede ser utilizada como un testigo preliminar para reconstruir la variación de la paleo-precipitación.

Palabras clave: Geoquímica, Relación elemental, Testigo de paleo-precipitación, Procedencia calcárea, Península de Yucatán.

VERSIÓN ABREVIADA EN CASTELLANO

Introducción

El calentamiento global antropogénico actual ha incrementado la curiosidad científica para comprender las respuestas de los diferentes ecosistemas, del régimen de precipitación y de la desertificación, ante el cambio climático experimentado en el pasado geológico (Masson-Delmotte et al., 2013). Los cambios estratigráficos en la fraccionación de isótopos estables, el contenido macro y micro fósil y la abundancia de los minerales en los archivos sedimentarios, han sido utilizados para reconstruir el ambiente del pasado y las condiciones paleo-climáticas (Curtis et al., 1996; Hodell et al., 2005; Carillo-Bastos et al., 2010; Torrescano-Valle e Islebe, 2015; Roy et al., 2016). La mayoría de los estudios paleoambientales realizados en la Península de Yucatán están basados en registros fosilizados de polen, pero poco se conoce sobre las concentraciones de elementos inorgánicos de los registros sedimentarios (Islebe y Sánchez, 2002; Carillo-Bastos et al., 2010; Aragón-Moreno et al., 2012; Gutiérrez-Ayala et al., 2012; Torrescano-Valle e Islebe, 2006, 2012). En este artículo se presentan una comparación de las concentraciones de K, Ti, Fe, Ca y Sr, registradas en 35 muestras de sedimento superficial del Holoceno tardío, colectadas en diferentes regímenes de precipitación a lo largo de la Península. La comparación y evaluación del efecto de la geología en la composición química de los sedimentos, permite, establecer un testigo de paleo-precipitación. La hipótesis de trabajo se basa en el hecho de que los minerales clásticos terrígenos son transportados desde el origen hacia la cuenca sedimentaria por escorrentía. Las concentraciones de K, Ti y Fe reflejan la abundancia de los minerales clásticos terrígenos, dicha concentración puede incrementar en sedimentos depositados durante los regímenes de alta precipitación.

Zona de estudio y muestreo

La Península de Yucatán (PY) se localiza en el sureste de México, es un lecho de roca caliza del Neógeno, con cantidades menores de lutitas del mismo periodo, también se encuentran areniscas y aluvión del Cuaternario (Servicio Geológico Mexicano, 2005a, 2005b; Fig. 1A). La PY recibe precipitación durante el verano y el otoño con una distribución bimodal y experimenta un periodo seco a mitad del verano llamado canícula (Magaña et al., 1999). Las masas de aire polar continental promueven lluvias durante el invierno y primavera aunque en menor cantidad (Mendoza et al., 2006). Se colectaron 35 muestras superficiales de sedimento provenientes de diferentes sitios de la Península de Yucatán de México (en los estados de Campeche, Yucatán y Quintana Roo; Fig. 1B). Estos sitios reciben una precipitación media anual entre 600 y 1600 mm. Las muestras fueron agrupadas en 5 grupos: 600-800 mm (número de muestras=2), 800-1000 mm (n=5), 1000-1200 mm/a (n=5), 1200-1400 mm (n=18) y 1400-1600 mm (n=5).

Materiales y métodos

Las muestras fueron secadas a 40°C y homogenizadas por medio de un mortero de ágata hasta la obtención de un polvo fino. Las concentraciones de K, Ti, Fe, Ca y Sr fueron medidas en un analizador marca Thermo Scientific Niton XL3t para medir la fluorescencia de rayos X (FRX). Además todos los elementos fueron medidos en un analizador convencional de FRX marca Siemens, en discos fusionados y prensados elaborados con material en forma de pellets de las 35 muestras secas y homogéneas. Los resultados del análisis FRX convencional muestran alta y positiva correlación ($r > 0.9$, $p < 0.05$) con los resultados obtenidos en Niton FRX (Fig. 2). Por medio de las líneas de regresión es posible corregir los datos de concentraciones obtenidos en el Niton XL3t, comparándolos con los resultados obtenidos de convencional FRX. Análisis estadísticos de correlación y conglomerados fueron realizados para identificar los comportamientos de los elementos.

Resultados y discusión

Concentración elemental vs. Geología

Los sedimentos superficiales analizados para la PY presentan las siguientes concentraciones: 0.91% (media=0.31%) de K, 0.01-0.76% (media=0.34%) de Ti, 0.01-3.12% (media=1.22%) de Fe, 0.40-37.70% (media=14.0%) de Ca y 27-4059 ppm (media=724 ppm) de Sr (Tabla 1). Ti y Fe muestran una distribución similar ($r=0.9$, $p<0.05$), la distribución de K es parcialmente comparable con las distribuciones de Ti y Fe ($r=0.5$, $p<0.05$). La distribución de Ca y Sr son comparables ($r=0.7$, $p<0.05$), ambos se relacionan negativamente con K, Ti y Fe ($r=0.6-0.8$, $p<0.05$). El primer grupo de elementos (K, Ti y Fe) está asociado con silicatos clásticos y aluminosilicatos, el segundo grupo de elementos (Ca y Sr) está presente en los carbonatos y evaporitas (Fig. 3).

Se observaron diferencias entre la composición química de los sedimentos colectados en sitios con geología similar. Las muestras colectadas en sitios con dominante contenido de calizas muestran diferentes abundancias de elementos asociados con minerales clásticos y carbonatos (Tabla 1). Las concentraciones de K, Ti y Fe son relativamente altas en los sedimentos superficiales de la PY, comparadas con la composición química de caliza promedio (Tabla 1; Mason y Moore, 1982), lo cual sugiere la influencia de rocas sedimentarias ricas en silicatos. A través de la erosión de las margas intercaladas en las calizas, areniscas y lutitas, los sedimentos superficiales recibieron cantidades variables de K, Ti y Fe.

Concentración elemental vs. Precipitación

La erosión y escorrentía ocurren en un ambiente de oxidación con pH y temperatura uniforme (Taylor y McLennan, 1985). Bajo estas condiciones, el potencial iónico controla la transportación de diferentes elementos, ya sea como material particulado o en el estado soluble (Mason y Moore, 1982). Las concentraciones de K, Ti y Fe reflejan abundancia de clastos terrígenos transportados por la escorrentía. Alta precipitación y escorrentía pueden depositar sedimentos con mayor concentración de K, Ti y Fe, y regímenes con baja precipitación pueden depositar menor cantidad de K, Ti y Fe. De forma similar, sedimentos depositados en regímenes de baja precipitación podrían tener mayor cantidad de Ca y Sr junto con la precipitación de carbonatos y evaporitas. En este trabajo se comparan concentraciones elementales de muestras superficiales de sedimento con la precipitación media anual (Fig. 4). Sin embargo, las concentraciones elementales son variables en cada régimen de precipitación y no presentan una clara tendencia con respecto a la precipitación media anual. Los sitios con precipitación media anual similar muestran diferentes concentraciones elementales. La presencia de marga y areniscas en conjunto con calizas posiblemente aportaron mayor concentración de K, Fe y Ti, en comparación con sitios constituidos predominantemente por calizas.

Relación elemental vs. Precipitación

Se utilizaron la relación entre elementos asociados con los minerales clásticos y elementos presentes en carbonatos y evaporitas (p.ej. K/Sr, Ti/Sr, Fe/Sr, K/Ca, Ti/Ca y Fe/Ca), para reducir el efecto de dilución por los carbonatos y evaporitas en la abundancia de minerales clásticos (p. ej. Weltje y Tjallingii, 2008). En la figura 5 se muestran los índices de variación elemental en sedimentos con respecto a la precipitación media anual de los sitios de muestreo. En general, los sedimentos con bajos valores de proporción elemental están presentes en sitios con menor precipitación media anual y no muestran una clara tendencia cuando la precipitación media anual incrementa. Estos valores pueden estar asociados a una baja escorrentía que aporta una mínima cantidad de clásticos y una menor erosión de los depósitos dominados por calizas (Tabla 1). Los sedimentos con menos clásticos (p. ej. K, Ti y Fe) no responden a la variable precipitación (Fig. 5). Sin embargo, se identificaron efectos intrínsecos del diseño de muestreo, por ejemplo, existe un mayor número de muestras ($n=18$) con precipitación entre 1200 y 1400 mm/a y un menor número de muestras ($n=2$) con precipitación entre 600 y 800 mm/a. Además se registró variación entre las cantidades de marga rica en clásticos y areniscas alrededor de los sitios de muestreo con regímenes de 1200-1400 mm/a y 1400-1600 mm/a que pueden contribuir a las variaciones en las abundancias de K, Fe y Ti. Para disminuir los efectos del muestreo y de la variable geológica del depósito, los valores promedio de K/Ca, Ti/Ca, Fe/Ca, K/Sr, Ti/Sr y Ti/Sr fueron comparados con la precipitación promedio anual. Las proporciones de K/Ca, Ti/Ca y Fe/Ca muestran tendencias positivas con la precipitación (Fig. 6). La proporción K/Ca es mejor que el Ti/Ca y Fe/Ca para inferir la variación de la paleo-precipitación en la Península, debido que Ti presenta baja abundancia y Fe puede ser móvil en ambientes anóxicos. Los resultados sugieren que los sedimentos con más clásticos responden mejor a la variación de precipitación y que la proporción K/Ca puede ser utilizada como un testigo preliminar para infe-

rir la paleo-precipitación en la Península. No obstante, se requiere incrementar el análisis de muestras de sedimento superficial provenientes de sitios con diferente régimen de precipitación a lo largo de la Península de Yucatán.

Conclusiones

Sedimentos que provienen de sitios con geología similar tienen composición química diferente. Además de la caliza dominante, la erosión de las lutita, marga y arenisca aportan diferentes cantidades de K, Ti y Fe.

La falta de relación entre las concentraciones elementales y la precipitación media anual es producto del efecto de dilución de carbonatos y evaporitas en los minerales clásticos.

Los valores promedio de K/Ca, Ti/Ca y Fe/Ca en sedimentos con alta abundancia de clásticos incrementan con respecto a un aumento en el valor de la precipitación media anual. La relación K/Ca es mejor que la de Ti/Ca y Fe/Ca, para inferir la variación de la paleo-precipitación en la Península de Yucatán. Sin embargo, se requiere incrementar muestras de sedimento provenientes de los sitios con bajo régimen de precipitación para poder evaluar la relación entre relación elemental y precipitación.

Introduction

Anthropogenic global warming of the modern era has increased scientific curiosity to document the responses of different ecosystems and regimes of precipitation and desertification to climate change over the geological past (Masson-Delmotte *et al.*, 2013). Stratigraphic changes of stable isotope fractionation, macro and micro fossil content and abundance of minerals in sedimentary archives have been used to infer past environmental and climatic conditions (Curtis *et al.*, 1996; Hodell *et al.*, 2005; Carillo-Bastos *et al.*, 2010; Torrescano-Valle and Islebe, 2015; Roy *et al.*, 2016). Elemental concentration indirectly estimates abundances of both the allochthonous and autochthonous minerals present in sediment and is less time consuming compared to the analysis of the micro-fossils and relatively less expensive compared to the analysis of stable isotopes. The chemistry of allochthonous fraction in sedimentary archives is affected by chemical weathering of clastic minerals during erosion and transportation from the source region and also by post-depositional alterations such as diagenesis and metasomatism (Nesbitt and Young, 1982; Mason and Moore, 1982; Taylor and McLennan, 1985; Cohen, 2003). However, it is possible to quantify these effects and select elements that best preserve a record of source rock erosion and transportation of terrigenous minerals by fluvial/pluvial activities in humid intervals and aeolian activity during arid intervals (Roy *et al.*, 2013; Roy *et al.*, 2015).

The pioneering studies of Haug *et al.* (2001, 2003) and Peterson and Haug (2006) presented Ti and Fe concentrations in sediments of the Cariaco Basin off Venezuela as a proxy to estimate riverine discharge and varying rainfall associated with the dynamics of

the Inter-Tropical Convergence Zone. Over the last decade, several researchers have successfully used concentrations of different elements in lacustrine sediments from distinct geological terrains to reconstruct the Late Quaternary paleoclimate of different parts of Mexico and infer the associated climatic forcings such as the North American Monsoon, El Niño Southern Oscillation, etc. (Metcalf *et al.*, 2010; Sosa-Najera *et al.*, 2010; Lozano-García *et al.*, 2015; Roy *et al.*, 2010, 2015, 2016). Compared to the rest of Mexico, chemical composition of sedimentary archives from the Yucatán Peninsula has received significantly less attention (Curtis *et al.*, 1996; Hodell *et al.*, 2005; Carillo-Bastos *et al.*, 2010). Most of the studies related to the paleoecology and paleoclimate of the peninsula are based on fossil pollen preserved in sediments of lacustrine basins as well as coastal swamps and marshes (Islebe and Sánchez, 2002; Carillo-Bastos *et al.*, 2010; Aragón-Moreno *et al.*, 2012; Gutiérrez-Ayala *et al.*, 2012; Torrescano-Valle and Islebe, 2006, 2012) and there is hardly any proxy register that involves inorganic element concentrations in sedimentary archives. In this paper, we present concentrations of K, Ti, Fe, Ca and Sr in 35 near-surface late Holocene sediment samples collected from different precipitation regimes across the Yucatán Peninsula. Elemental concentrations of sediments are compared with mean annual rainfall of the sampling sites in order to establish a proxy for precipitation after evaluating the effect of geology. Our working hypothesis is based on the fact that terrigenous clastic minerals are transported from the catchment area into the sedimentary basins by runoff. Concentrations of K, Ti and Fe would reflect abundance of the terrigenous clastic minerals and their concentrations would increase in sediments deposited during regimes of higher precipitation.

Study area and sampling

The Yucatán Peninsula is located in southeastern Mexico and its bedrock has dominant Neogene limestone and minor amounts of Neogene shale and Quaternary sandstone and alluvium (Servicio Geológico Mexicano, 2005a, 2005b; Fig. 1A). It receives precipitation during the summer and autumn months in a bimodal distribution with a mid-summer dry spell (Magaña *et al.*, 1999). The polar continental air masses bring minor winter and spring precipitations (Mendoza *et al.*, 2006). A total of 35 surface sediments were collected from different sites across the peninsula (i.e. the states of Campeche, Yucatán and Quintana Roo, Fig. 1A) from permanent (e.g. lakes, swamps) as well as seasonal water bodies. The sampling sites receive mean annual precipitation of between 600 and 1,600 mm (INEGI, 2006; Fig. 1B). We divided the sampling sites into groups with different amounts of mean annual precipitation: 600-800 mm (n=2), 800-1,000 mm (n=5), 1,000-1,200 mm (n=5), 1,200-1,400 mm (n=18) and 1,400-1,600 mm (n=5). More than half of the sampling sites are within the precipitation group of 1,200-1,400 mm due to the relatively easier access to these sampling sites as well as the location of most of the water bodies being in higher precipitation regimes.

Material and methods

Surface sediments comprise organic-rich clay as well as calcareous clay and some of them contain abundant root remnants and coarser sand and gravel (Table 1). Samples were oven dried at 40°C and sieved to remove the root remnants and coarser fragments and subsequently homogenized and ground with an agate mortar and pestle. Concentrations of K, Ti, Fe, Ca and Sr were measured in dry and powdered samples using the portable Thermo Scientific Niton XL3t X-ray fluorescence (XRF) and non-destructive sample preparation technique. Analysis of three different geological reference materials (OU-8, BCU-3 and IGLA-1) suggests an error of 1-6% in precision and an error of 1.5-392% in accuracy. Concentrations of K in 7 samples, Ti in 11 different samples and Fe in one sample remained below the detection limit of the Niton XRF equipment and hence could not be measured. We measured all the elements again in a conventional Siemens XRF by making a fused disc and a pressed pellet from dry powdered sediments. Approximately 0.8 g of dry and powder sediment was mixed with 7.2 g of (1:1) LiBO₂ and Li₂B₄O₇ mixture and this mixture was fused in a Pt/Au (95:5) alloy crisol to analyze the concentrations of K, Ti, Fe and Ca as per Lozano-Santacruz *et al.* (1995). The concentration of K in one

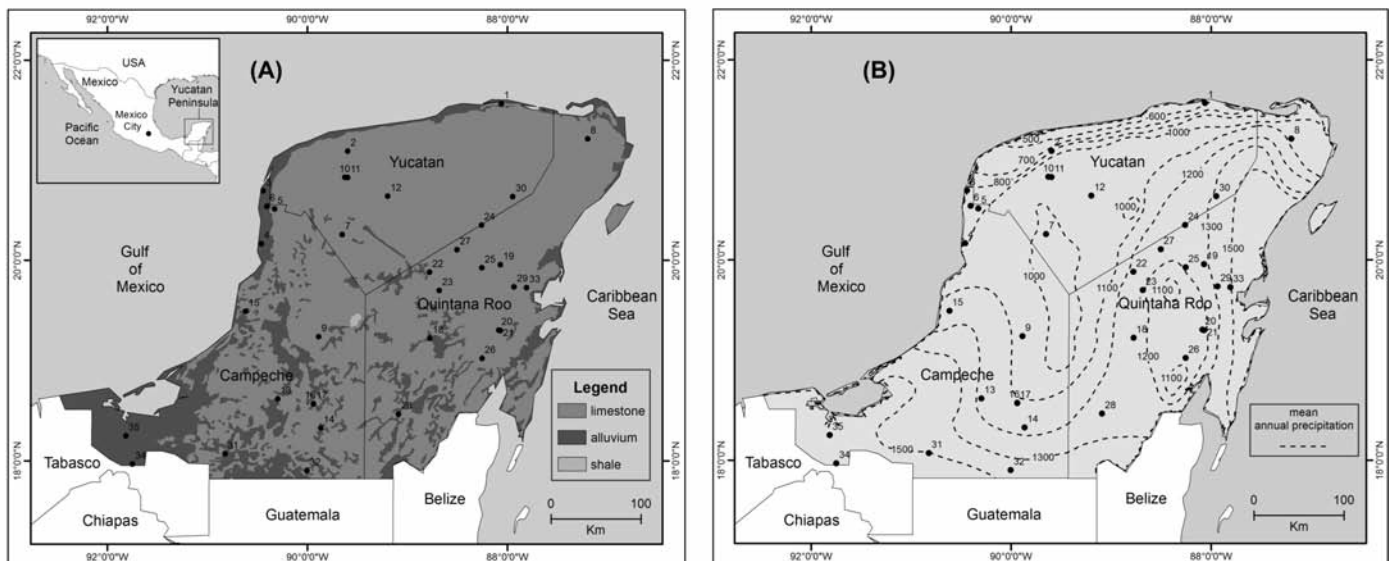


Figure 1. Location of sampling sites in (A) different geological provinces and (B) precipitation regimes of the Yucatán Peninsula of Mexico. Samples were collected from sites with catchments dominantly comprising of limestone. The mean annual precipitation varies between 600 mm and 1,600 mm.

Figura 1. Localización de los sitios de muestreo en (A) diferentes provincias geológicas y (B) regímenes de precipitación de la Península de Yucatán. Las muestras fueron colectadas en sitios con afloramiento dominante de caliza. La precipitación media anual varía entre 600 y 1600 mm.

Mean precipitation	Sample	Location (Latitude, Longitude)	K (%)	Ti (%)	Fe (%)	Ca (%)	Sr (ppm)	Sediment texture
600-800 mm/a	1	21° 33' 60" N, 88° 03' 34.3" W	0.06	0.02	0.08	27.20	2748	clay/calcareous
	2	21° 5' 21.1" N, 89° 35' 44.6" W	0.83	0.46	2.18	5.09	105	sand/gravel/organic
800-1000 mm/a	3	20° 41' 33.1" N, 90° 26' 25.9" W	0.01	<lod	0.05	26.35	2334	clay/calcareous
	4	20° 09' 48.4" N, 90° 27' 31.1" W	0.04	0.01	0.05	22.56	4059	clay/calcareous
	5	20° 30' 47.2" N, 90° 19' 35.2" W	0.04	0.03	0.16	7.71	635	clay/organic
	6	20° 32' 22.4" N, 90° 24' 11.1" W	0.02	<lod	0.05	22.35	1161	clay/organic
	7	20° 15' 18.3" N, 89° 39' 2.3" W	0.37	0.34	1.56	11.50	36	clay/organic/root
1000-1200 mm/a	8	21° 12' 41.9" N, 87° 11' 41.8" W	0.01	0.02	0.05	34.49	532	clay/calcareous/fossil
	9	19° 14' 29.8" N, 89° 53' 7.1" W	0.68	0.37	2.02	2.35	37	clay/root/organic
	10	20° 49' 44.3" N, 89° 37' 31.0" W	0.39	0.33	1.49	5.33	117	clay/sand/root
	11	20° 49' 31.4" N, 20° 35' 44.6" W	0.06	0.08	0.27	30.27	113	clay/organic/root
	12	20° 38' 26.1" N, 89° 11' 45.7" W	0.43	0.25	1.26	10.85	121	sand/fossil
1200-1400 mm/a	13	18° 37' 10.0" N, 90° 17' 43.9" W	0.05	0.19	0.97	16.28	406	sand/gravel/organic
	14	18° 19' 40.2" N, 89° 51' 56.2" W	0.91	0.35	1.77	5.86	81	clay/calcareous
	15	19° 29' 42.4" N, 90° 36' 56.3" W	0.30	0.76	2.49	0.93	38	clay/gravel/calcareous
	16	18° 34' 21.7" N, 89° 56' 18.3" W	0.62	0.40	1.96	1.96	36	clay/root
	17	18° 34' 13.5" N, 89° 56' 18.8" W	0.58	0.26	1.42	11.21	34	clay/organic/root
	18	19° 13' 30.2" N, 88° 46' 28.0" W	0.47	0.61	1.24	0.96	37	clay/calcareous
	19	19° 57' 21.3" N, 88° 04' 7.5" W	0.00	<lod	0.01	36.34	2571	clay/root/calcareous
	20	19° 17' 87.2" N, 88° 05' 09.2" W	0.01	0.04	0.01	34.63	1606	clay/root/calcareous
	21	19° 18' 03.1" N, 88° 04' 11.3" W	0.00	<lod	0.02	35.34	2313	clay/root/organic/calcareous
	22	19° 52' 45.1" N, 88° 46' 36.0" W	0.28	0.15	0.73	18.56	1923	clay/gravel/calcareous
	23	19° 41' 46.7" N, 88° 40' 50" W	0.68	0.62	2.90	1.07	27	silt/root/organic
	24	20° 20' 55.3" N, 88° 15' 28.1" W	0.47	0.55	2.68	2.71	93	clay/gravel/root
	25	19° 55' 28.6" N, 88° 15' 13.6" W	0.47	0.30	1.49	8.14	101	clay/gravel/root
	26	19° 01' 22.9" N, 88° 15' 11.7" W	0.31	0.23	1.15	16.71	246	clay/gravel/organic/calcareous
	27	20° 06' 15.6" N, 88° 30' 07.9" W	0.67	0.63	2.95	2.58	50	clay/gravel/root
	28	18° 28' 1.8" N, 89° 05' 20.5" W	0.54	0.32	1.35	8.07	28	silt/gravel/fossil/organic
	29	19° 44' 0.4" N, 87° 55' 59.2" W	0.00	<lod	0.03	35.84	1218	clay/calcareous
	30	20° 38' 05.4" N, 87° 56' 46.3" W	0.36	0.69	3.12	1.23	56	silt/root
1400-1600 mm/a	31	18° 04' 12.6" N, 90° 49' 22.7" W	<lod	<lod	0.02	37.70	1867	clay/root/calcareous
	32	17° 54' 7.7" N, 90° 00' 10.3" W	0.05	0.43	2.24	1.60	73	clay/root
	33	19° 43' 33.4" N, 87° 48' 23.2" W	0.13	0.47	2.08	2.60	412	silt/gravel/root
	34	17° 58' 6.9" N, 91° 44' 54.5" W	0.32	0.55	1.13	0.65	88	silt/root/organic
	35	18° 15' 5.5" N, 91° 48' 53.1" W	0.22	0.53	1.60	0.39	52	silt/root/organic
	UCC		2.80	0.30	3.50	3.00	350	
	limestone		0.14	0.04	0.19	30.39	-	

<lod; below limit of detection, UCC; average upper continental crust composition (Taylor and McLennan, 1985), limestone; average limestone composition (Mason and Moore, 1982)

Table 1. Location, chemical composition and texture of samples collected from sites with different mean annual precipitation across the Yucatan Peninsula, Mexico.

Tabla 1. Ubicación, composición química y textura de las muestras colectadas en sitios con diferente precipitación media anual a lo largo de la Península de Yucatán, México.

sample and Ti in six samples remained below the detection limit of the equipment. The concentration of Sr was measured in the pressed pellets prepared from the mixture of 4 g of powder sediment and binder following the instructions of Lozano-Santacruz *et al.* (1995) and Verma *et al.* (1996). Loss on ignition was estimated by a gravimetric method after heating 1 g of sample up to 950 °C in a porcelain crucible for 1 hour. The results obtained in the conventional XRF exhibit strong positive correlations ($r > 0.9$, $p < 0.05$) with the results obtained in Niton XL3t XRF (Fig. 2). Concentrations of K, Fe and Ca were overestimated and concentration of Sr was underestimated in Niton XL3t compared to conventional Siemens XRF. Both types of XRF equipment provided almost similar concentrations of Ti. It was possible to minimize the differences between concentrations obtained with both

the XRF spectrometers by using linear regression obtained after comparing results from both. However, we used the results obtained in conventional XRF for interpretation as many samples analyzed in Niton XL3t XRF lacked concentrations of K, Ti and Fe and hence could not be corrected. Statistical analysis (correlation and cluster) was performed using the STATISTICA software to identify the associations of different elements.

Results and discussion

Elemental concentration vs. geology

Geology controls the chemical composition of sediments as elements associated with terrigenous clastic

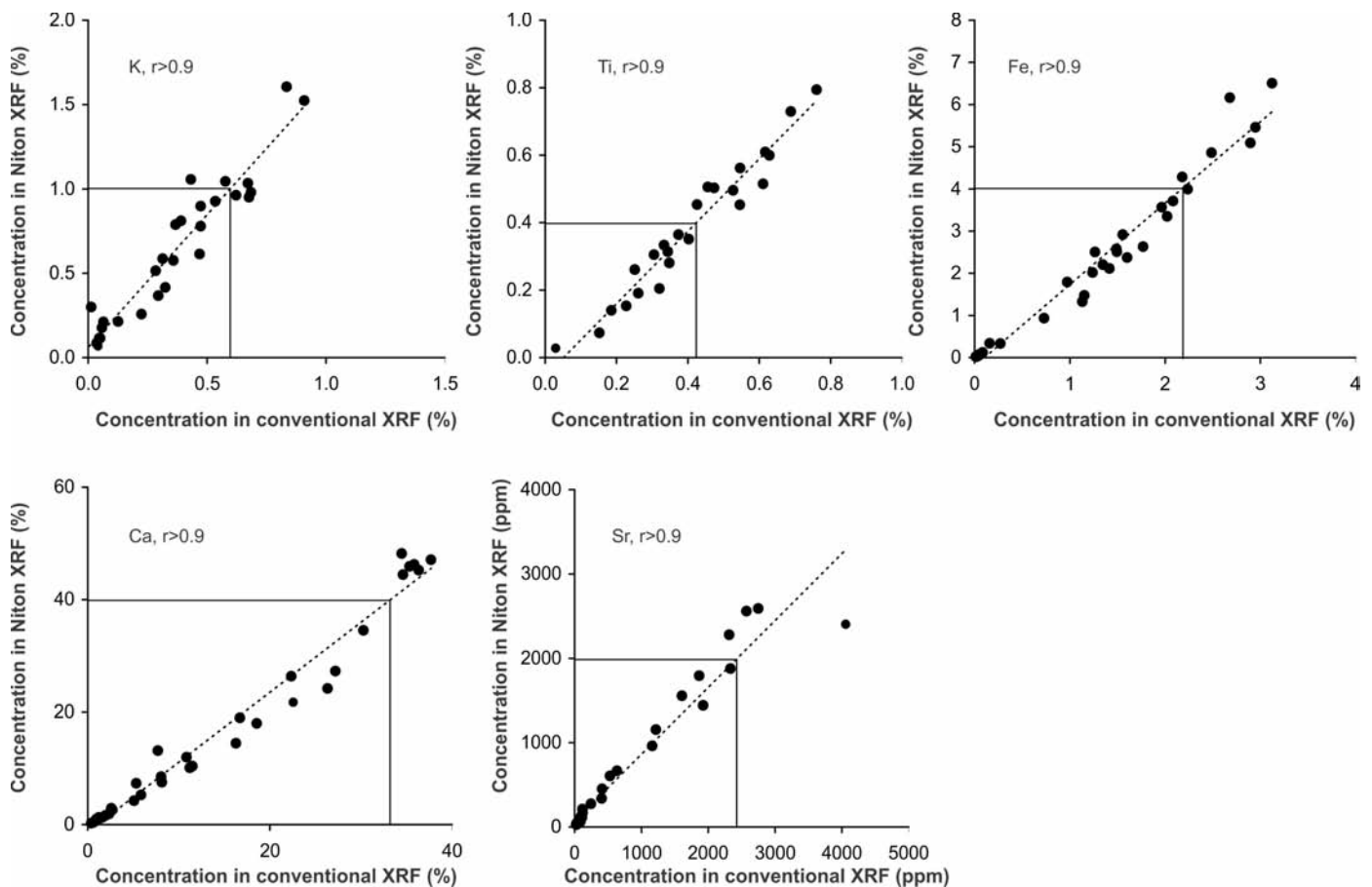


Figure 2. Comparison of concentrations of K, Ti, Fe, Ca and Sr obtained in Niton XL3t XRF and traditional Siemens XRF in near-surface sediments of the Yucatán Peninsula. Concentrations of K, Fe and Ca were overestimated and Sr is underestimated in Niton XRF compared to the traditional XRF.

Figura 2. La comparación de las concentraciones de K, Ti, Fe, Ca y Sr obtenidas por medio de un equipo Niton XL3t XRF y un equipo tradicional Siemens XRF, en sedimentos de la Península de Yucatán. Las concentraciones de K, Fe y Ca fueron sobre-estimadas y el Sr fue sub-estimado en el equipo Niton XL3t XRF en comparación con el equipo tradicional.

minerals (silicates and aluminosilicates) are derived from erosion of the catchment rocks and elements associated with authigenic minerals (carbonates and evaporites) are deposited from the soluble ions present in water column (Mason and Moore, 1982; Taylor and McLennan, 1985; Cohen, 2003). Surface sediments from the Yucatán Peninsula have up to 0.91% (mean=0.31%) of K, 0.01-0.76% (mean=0.34%) of Ti, 0.01-3.12% (mean=1.22%) of Fe, 0.40-37.70% (mean=14.0%) of Ca and 27-4059 ppm (mean=724 ppm) of Sr (Table 1). Ti and Fe show similar ($r=0.9$, $p<0.05$) distributions and the distribution of K is comparable partly to the distributions of Ti and Fe ($r=0.5$, $p<0.05$). Similarly, the distributions of Ca and Sr are comparable ($r=0.7$, $p<0.05$) and both of them are negatively related to K, Ti and Fe ($r=0.6-0.8$, $p<0.05$). The first group of elements (K, Ti and Fe) is associated with clastic silicates and aluminosilicates and the second group of elements (Ca and Sr) is present in carbonate and evaporites (Fig. 3).

Less K and Fe in all the samples and higher Ca and Sr in many of the samples were not similar to the average composition of the Upper Continental Crust (UCC; Taylor and McLennan, 1985) and thus reflect the influence of the regional geology. The geology of the Yucatán Peninsula of Mexico is characterized by

abundant limestone and the alluvium produced from erosion of the carbonate-rich lithology (Servicio Geológico Mexicano, 2005a, 2005b). However, we observed dissimilarities between chemical compositions of sediments collected from the sites with similar catchment geology. Samples collected from sites with dominant limestone show different abundances of elements associated with both clastics minerals and carbonates. For example, samples 2, 7, 8 and 26 were collected from sites with dominant limestone and all of them have different concentrations of K, Ti and Ca (Table 1). Sample 2 has higher K (0.83%) and Ti (0.46%) and lower Ca (5.09%) compared to sample 8 (K: 0.01%; Ti: 0.02%; Ca: 34.49%). Even the samples (3, 4, 34 and 35) collected from the alluvium-rich catchment geology have different abundances of K, Ti and Ca. Some samples have more K and Ti and others have more Ca.

Apart from the two major lithologies (i.e. limestone and alluvium), the geology of the Yucatán Peninsula is also characterized by presence of marl in limestone in both the Carillo Puerto Formation and Chichen Itza Formation (Servicio Geológico Mexicano, 2005a, 2005b). Additionally, shale and poorly consolidated sandstones are exposed in different parts of the peninsula (Servicio Geológico

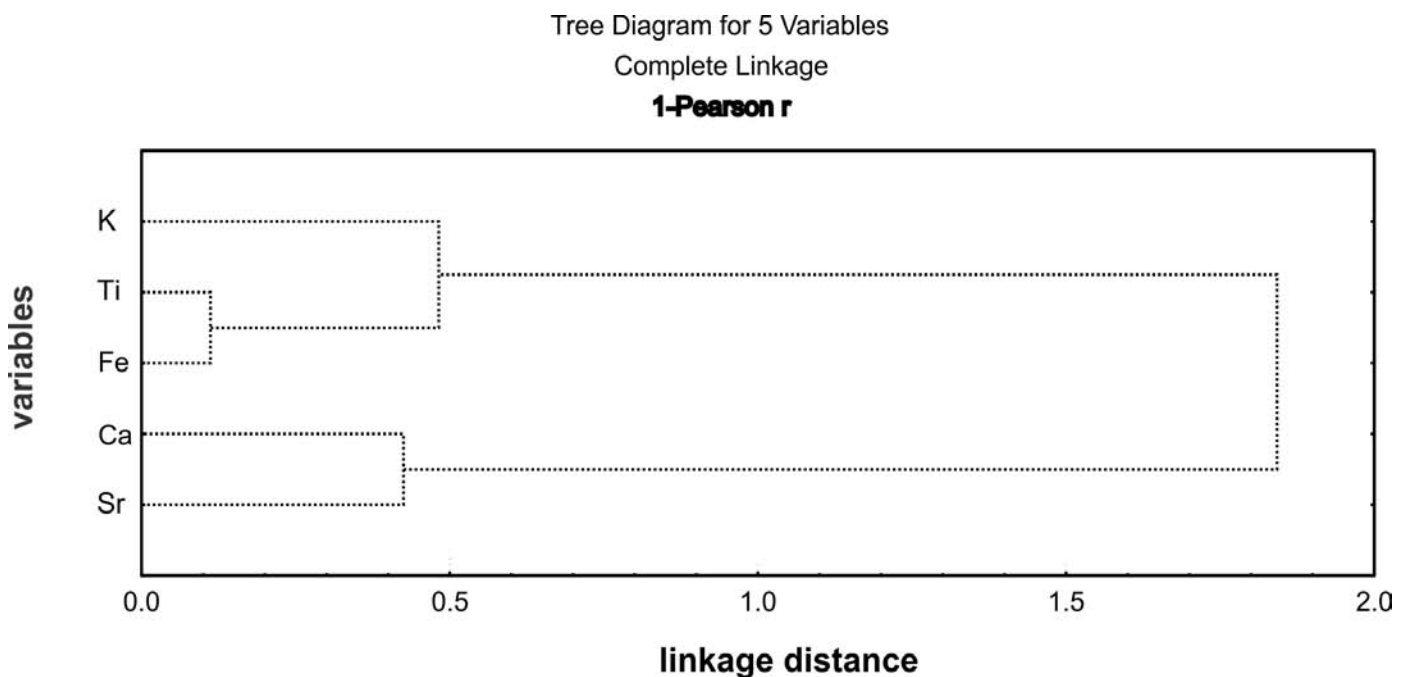


Figure 3. Tree-diagram of measured elements in surface sediments of the Yucatán Peninsula based on 1-Pearson-r and complete linkage methods. Association of K, Ti and Fe represents the clastic minerals and association of Ca and Sr represents carbonates and evaporites.
Figura 3. El diagrama de árbol de los elementos medidos en sedimentos superficiales de la Península de Yucatán, está basado en el método 1-Pearson-r y el de enlace completo. La asociación de K, Ti y Fe representa los minerales clásticos y la asociación de Ca y Sr representa los carbonatos y evaporitas.

Mexicano, 2005a, 2005b). Relatively higher concentrations of K, Ti and Fe in near-surface sediments of the Yucatan Peninsula compared to the average limestone (Table 1; Mason and Moore, 1982) suggesting the influence of these clastic-rich sedimentary rocks. Erosion of different amounts of marl as well as sandstone and their different contributions in different parts of the peninsula led to variable amounts of K, Ti and Fe being found in the surface sediments.

Elemental concentration vs. precipitation

Erosion of catchment rocks and subsequent transpor-

tation of terrigenous clastic sediments by runoff occurs in a generally oxidizing environment with relatively uniform pH and temperature (Taylor and McLennan, 1985). Under such conditions, ionic potential controls transportation of different elements, either as particulate matter (oxides and association with hydroxyl groups) or in soluble state (Mason and Moore, 1982). Compared to soluble alkali (K) and alkaline earth (Ca and Sr) elements, the quadrivalent Ti and trivalent Fe remain immobile and insoluble. Similarly, the early soluble K subsequently adsorbs onto the finer clastic fractions and clay minerals due to its larger ionic radius. Thus, concentrations of K, Ti and Fe reflect the abundance of the terrigenous clas-

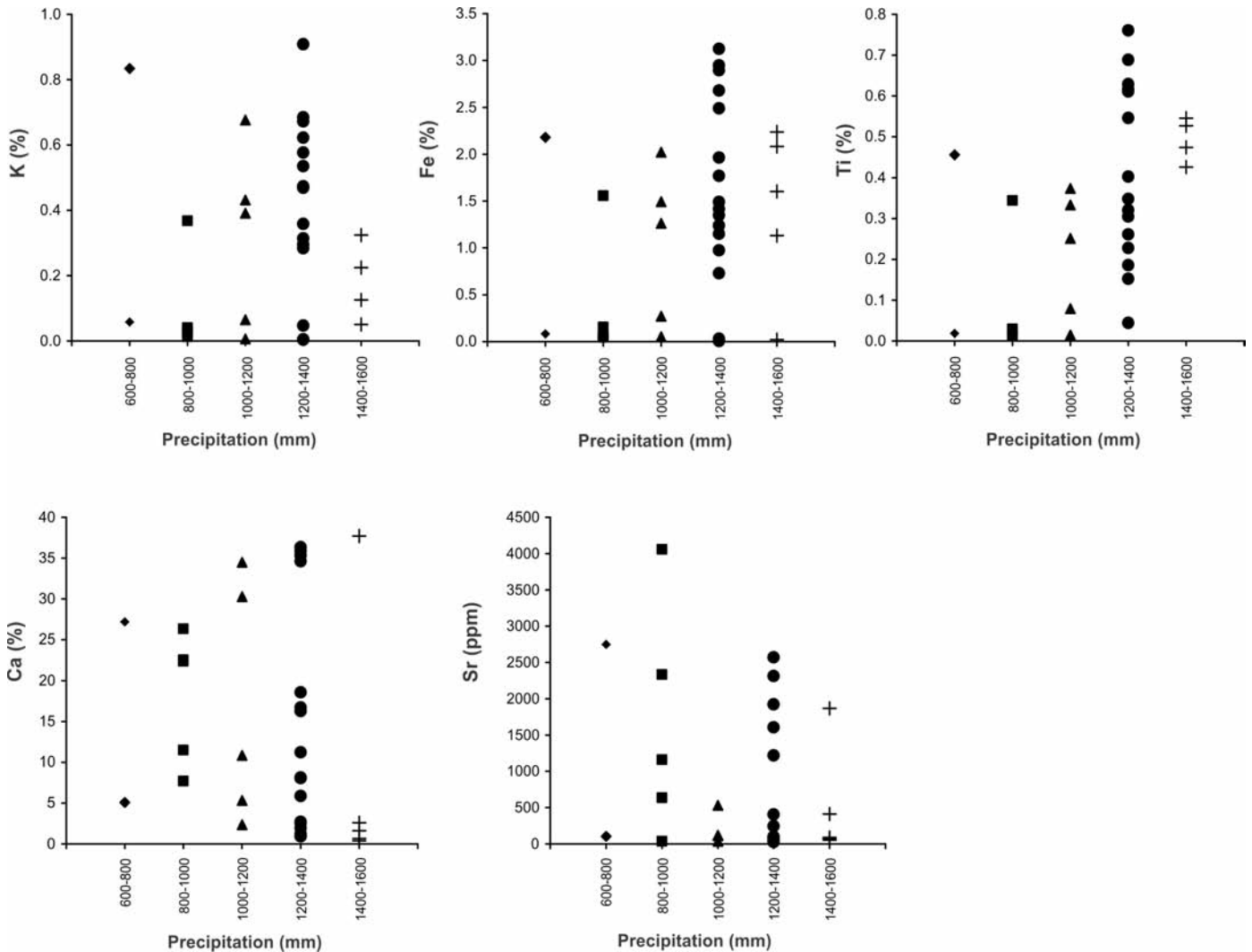


Figure 4. Binary plots showing concentrations of K, Ti, Fe, Ca and Sr in near-surface sediments and mean annual precipitation of the sampling sites.

Figura 4. Los gráficos binarios muestran las concentraciones de K, Ti, Fe, Ca y Sr en sedimentos superficiales y la precipitación media anual de los sitios de muestreo.

tic minerals transported by runoff. Higher precipitation and more runoff would deposit sediments with more K, Ti and Fe and regimes of lower precipitation would deposit sediments with less K, Ti and Fe. Similarly, sediments deposited in regimes of lower precipitation would have more Ca and Sr as the soluble alkaline earth metals would deposit by precipitation of carbonates and evaporites.

We compared elemental concentrations of the surface sediments with mean annual precipitation of the sampling sites (Fig. 4) and expected to observe higher values of K, Ti and Fe and lower Ca and Sr in sites with more annual precipitation compared to the sites that

receive less annual precipitation. However, elemental concentrations are variable and do not show any clear trend with respect to precipitation. The concentration of K is 0.01% in one sample and 0.91% in another sample and both the samples were collected from locations that receive 1200-1400 mm of mean precipitation. Similarly, Fe is 0.05% in one sample and 2.02% in another sample collected from sites located in a region receiving 1000-1200 mm of rainfall. Samples with lower K and Fe have higher Ca and Sr and vice versa. As the elemental concentrations are expressed in relative abundance (i.e. weight percentage), variable concentration of elements represen-

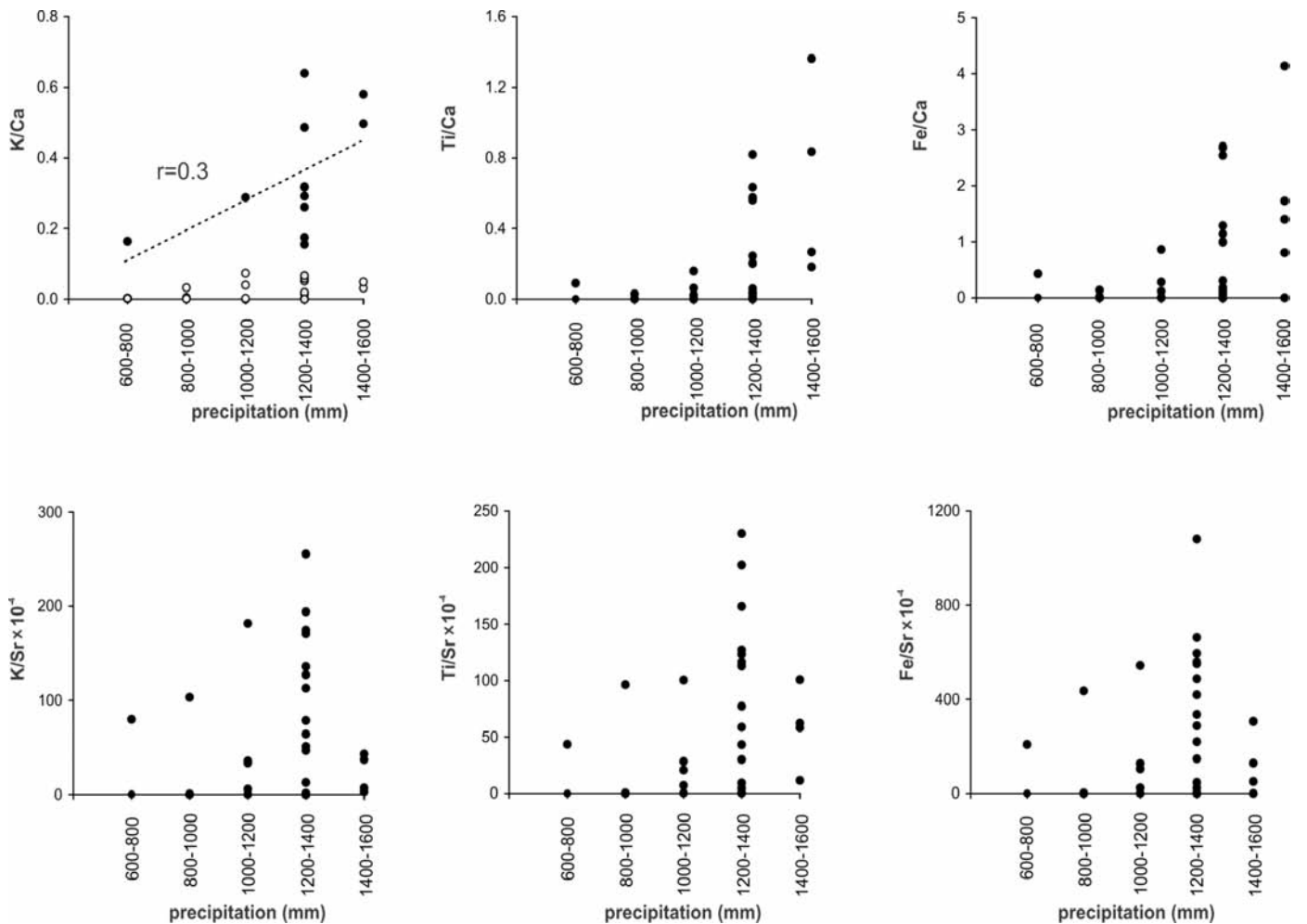


Figure 5. Binary plots showing elemental ratios of near-surface sediments and mean annual precipitation of the sampling sites. Samples with minimal clastics (open circles) exhibit lower and homogeneous element ratios irrespective of the amount of mean annual precipitation. Samples with more clastic minerals (filled circles) have variable values.

Figura 5. Los gráficos binarios muestran la relación elemental de los sedimentos superficiales y la precipitación media anual de los sitios de muestreo. Las muestras con un mínimo de clastos (círculos abiertos) exhiben proporciones de elementos más bajas y homogéneas, independientemente de la cantidad de la precipitación media anual. Las muestras con más minerales clásticos (círculos llenos) tienen valores altamente variables.

ting the clastic mineral abundance from locations with comparable mean annual precipitation is a reflection of different degrees of dilution caused by the carbonates and evaporites. The cluster of elements associated with carbonates and evaporites exhibit negative correlations with the cluster of elements present in the terrigenous clastic minerals. Additionally, sediments sourced from catchments with minor amounts of marl and sandstone along with dominant limestone can have relatively higher concentrations of K, Fe and Ti compared to samples sourced from catchments consisting of only limestone, although both the groups of sediments are collected from sites with comparable mean annual precipitation.

Elemental ratio vs. precipitation

We used ratios of elements associated with clastic minerals and elements present in carbonates and evaporites (i.e. K/Ca, Ti/Ca, Fe/Ca, K/Sr, Ti/Sr and Fe/Sr) in order to minimize the dilution effect of carbonates and evaporites on the abundance of clastic minerals (e.g. Weltje and Tjallingii, 2008). Figure 5 shows the variation of elemental ratios in sediments with respect to mean annual precipitation. In general, the sediments with lower and homogenous values of elemental ratios are present in locations with less mean annual precipitation and they do not show any clear tendency as the mean annual precipitation increases. The lower values can be due to minimal abundances

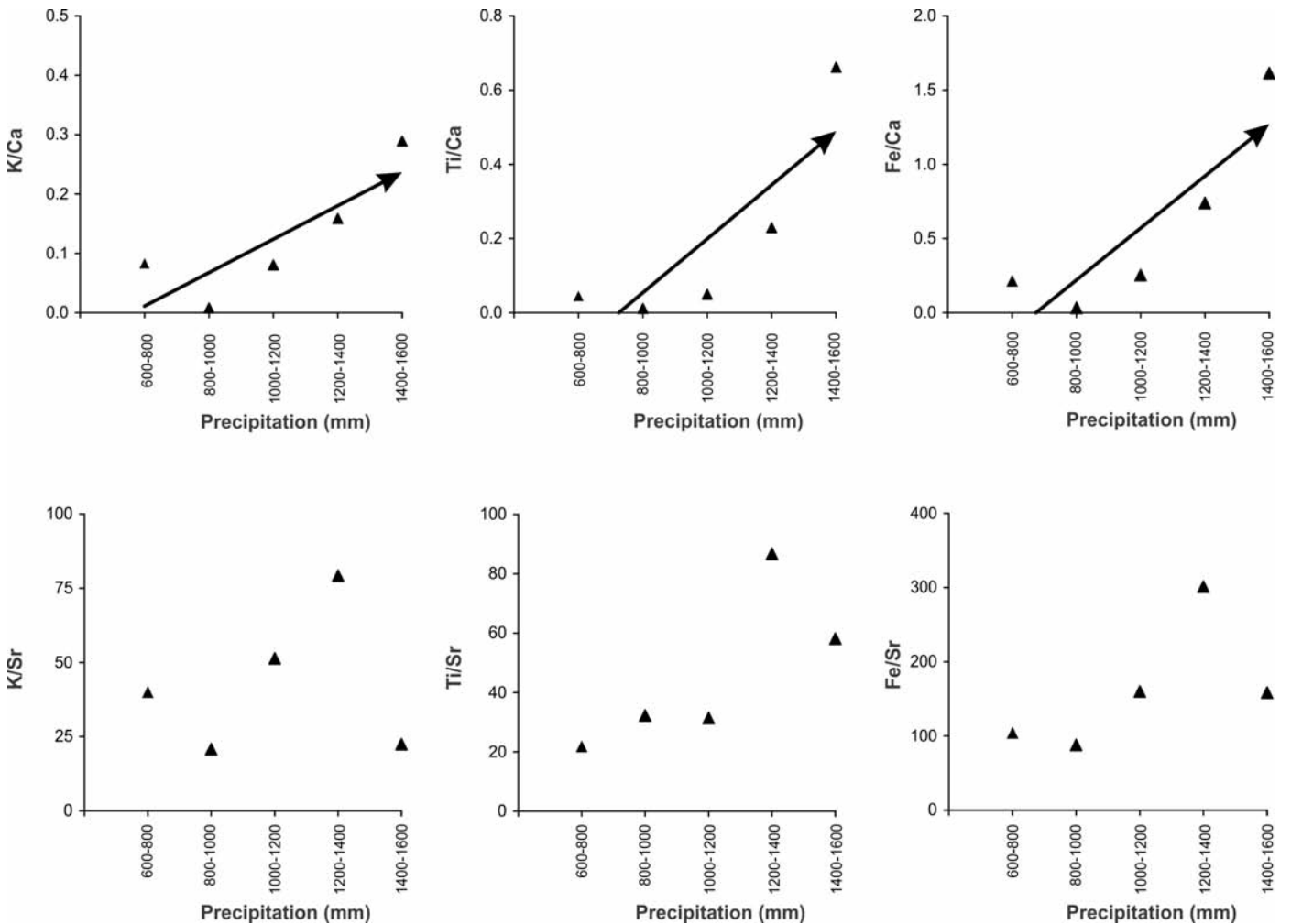


Figure 6. Tendencies of average values of K/Ca, Ti/Ca, Fe/Ca, K/Sr, Fe/Sr and Ti/Sr with respect to mean annual precipitation of the sampling sites.

Figura 6. Tendencias de los valores promedio de K/Ca, Ti/Ca, Fe/Ca, K/Sr, Fe/Sr y Ti/Sr con respecto a la precipitación media anual de los sitios de muestreo.

of clastics as a result of less runoff into these sites as well as limestone-dominated catchments (Table 1). Sediments with minimal clastic minerals (i.e. less K, Ti and Fe) do not respond to the variable precipitation (open circles in Fig. 5). Among the sediments with a higher abundance of clastic minerals, the samples that were collected from locations with higher mean annual precipitation generally exhibited a higher element ratio compared to the sediments collected from sites with lower mean annual rainfall (filled circles in Fig. 5). However, the samples from the precipitation group of 1,200-1,400 mm have variable values of K/Ca and samples from the groups of 1,200-1,400 mm and 1,400-1,600 mm have variable values of Ti/Ca and Fe/Ca. Some of these values are similar or even lower than samples collected from other sites with relatively less mean annual precipitation. Biased sampling, as well as different composition of catchment geology in sites of comparable isohyets, may be the reasons. The number of samples from sites with mean annual precipitation of 1,200-1,400 mm is much higher ($n=18$) compared to the samples collected from sites located within other precipitation groups ($n=2-5$). Similarly, the varying amounts of clastic-rich marl and sandstone around the sampling sites with 1,200-1,400 mm and 1,400-1,600 mm of annual precipitation possibly contributed different abundances of K, Fe and Ti-bearing clastic sediments and hence the variable elemental ratios. We also observed that element/Ca ratios are better surrogates of precipitation compared to the element/Sr ratios as K/Sr, Ti/Sr and Fe/Sr are lower in sediments collected from locations of higher mean annual precipitation (i.e. 1,400-1,600 mm) compared to sites that receive relatively less precipitation (i.e. 1,200-1,400 mm).

In order to minimize the effects of biased sampling and variable catchment geology, the average values of K/Ca, Ti/Ca, Fe/Ca, K/Sr, Ti/Sr and Fe/Sr are compared with the mean annual precipitation (Fig. 6). Average values of the ratios of K/Ca, Ti/Ca and Fe/Ca show positive trends with respect to the precipitation. Sediments collected from sites with higher mean annual precipitation have higher ratios. However, this is a preliminary observation and further research is being carried out by increasing the number of samples as well as by improving the sample distribution in different precipitation regimes across the peninsula. Among the elemental ratios, K/Ca ratio is a better proxy of precipitation as calcareous-sediments from the Yucatán Peninsula have lower concentration of Ti and Fe can be mobile in anoxic depositional environments. Many surface samples from the limestone-rich Yucatán Peninsula have Ti below the detection limit of portable Niton XRF spectrometer and even the con-

ventional XRF could not measure Ti in six different samples. Similarly, the oxidation-reduction potential (Eh) can change in the post-deposition environment and lead to anoxic condition. Under such conditions, the insoluble Fe changes oxidation state and become mobile and soluble (Taylor and McLennan, 1985) and subsequently deposit as Fe-carbonate (e.g. siderite) or Fe-sulphide (e.g. pyrite) (Aller *et al.*, 1986; Suits and Wilkin, 1998). Anoxic conditions near the sediment-water interface of the Amazon River mouth led to presence of dissolved Fe and elevated HCO_3^- and caused the deposition of siderite (Aller *et al.*, 1986). Suits and Wilkin (1998) reported pyrite formation in the water column of the Green Lake (near Fayetteville, New York) as a result of euxinic conditions. Similarly, Sheu and Presley (1986) observed iron sulfides in sediment from the anoxic Orca Basin of the Gulf of Mexico. Previous research on sub-surface sediments from different parts of the peninsula reports higher organic carbon contents (10-30%, Leyden *et al.*, 1996; 15-25%, Gutiérrez-Ayala *et al.*, 2012) and the possible existence of anoxic depositional environments. In such conditions, the Fe present in the sediments would not indicate the abundance of clastic minerals.

Conclusions

Most of the paleoecology and paleoclimate research in the Yucatán Peninsula of Mexico is based on fossil pollen in sediments of lakes, coastal swamps and marshes and there is hardly any proxy register that involves inorganic element concentrations in sedimentary archives. In this paper, we present concentrations of five different elements (K, Ti, Fe, Ca and Sr) in 35 near-surface late Holocene sediment samples collected from different precipitation regimes across the peninsula. Elemental concentrations are compared with mean annual precipitation of the sampling sites in order to establish a proxy for precipitation after evaluating the effect of geology on sediment chemical composition. The main conclusions of this study are;

1. Sediments from sites with a similar catchment geology have dissimilar chemical compositions. Apart from the dominant limestone, the erosion of clastic-rich shale, marl and sandstone and their different contributions in different parts of the peninsula led to variable amounts of K, Ti and Fe.

2. Dilution effect of carbonates and evaporites on the clastic mineral abundance caused the lack of relationships of individual elemental concentrations and elemental ratios of sediments with minimal clastics with the mean annual precipitation.

3. Element/Ca ratios are better surrogates for precipitation compared to the element/Sr ratios. In general, the average values of K/Ca, Ti/Ca and Fe/Ca in sediments with higher clastic abundance increase with respect to increasing mean annual precipitation. However, the sampling protocol and presence of most of the sampled water bodies being in a single precipitation cluster (i.e. 1,200-1,400 mm/year) did not allow a complete evaluation of the potential correlation between precipitation and elemental ratios in near-surface sediments of the Yucatán Peninsula. In a preliminary observation, the K/Ca ratio could be used as a proxy to infer paleo-precipitation as sedimentary records in the karstic terrain have low Ti and Fe is mobile in anoxic environments.

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