

GEOQUÍMICA ORGÀNICA
DE CONQUES LACUSTRES FÒSSILS

CONCLUSIONS

S'ha dut a terme un extens estudi de la fracció lipídica de la matèria orgànica sedimentària d'una sèrie representativa de conques lacustres fòssils, com les de Ribesalbes, Campins, Libros, Rubielos i Cerdanya. Aquest ha inclòs l'anàlisi entre d'altres, d'hidrocarburs, alcohols, aldèhids, cetones, àcids, hidroxiaïcids, tiolans, tians i tiofens, amb estructura lineal, ramificada (p.e: isoprenoides) o policíclica (esterans, hopans, oleanans, ursans i lupans).

Un primer resultat d'aquest treball analític ha consistit en la descripció de nous compostos o noves sèries homòlogues de compostos no trobats prèviament en mostres geològiques, que ha permès d'extendre el nombre de biomarcadors o marcadors moleculars en geoquímica orgànica. Així:

a) S'han trobat sèries de 2-alcanols, 3-alcanols, 4-alcanols, 5-alcanols i 6-alcanols, que comprenen distribucions entre C23 i C33 amb predomini dels homòlegs de longitud de cadena senar. Aquestes són paral.leles a les distribucions de n -alcans sedimentaris provinents de les ceres cuticulars de les plantes superiors i molt probablement són originades per oxidació microbiana d'aquests compostos. En treballs anteriors a la present memòria de tesi, els únics alcohols lineals descrits en medis sedimentaris consistien en distribucions de 1-alcanols, i en alguns casos, de 2-alcanols corresponents a aportos algals i microbians específics.

b) S'han identificat distribucions d'etil, propil, butil i pentilcetones que comprenen un rang entre C23 i C33 amb predomini dels homòlegs de nombre de carboni senar. Aquestes distribucions són paral.leles a les de n -alcans característics de les ceres de plantes superiors i a les de metilcetones generades per oxidació microbiana dels anteriors, que fins ara constituïen les úniques mesclures conegudes de cetones lineals en medis sedimentaris. La semblança entre aquestes distribucions i les característiques dels ambients deposicionals on aquestes cetones s'han trobat, són indicatives d'un origen per retreballament microbià dels n -alcans sedimentaris.

c) S'han identificat distribucions modals de n -aldèhids entre C14 i C28 sense predomini parell-senar i màxim entorn del n -docosanal. Aquestes són complementàries a les distribucions de n -aldèhids entre C24 i C30 amb fort predomini dels de nombre de carboni parell, provinents de les plantes superiors, recentment descrites. Les sèries de n -aldèhids mostrades a la present memòria de tesi indiquen que aquests compostos no poden ésser sistemàticament atribuïts a aportos terrestres, sinó també a contribucions bacterianes.

d) Dins la fracció d'alcanodiols analitzada, s'han identificat per primera vegada els 1,13-, 14-, 16- i 17-triacontandiols, els 1,13- i 14-hentriacontandiols i el 1,19-hexatriacontandiòl.

e) Paral·lelament al cas anterior, dins de les mescles complexes de cetoalcohols analitzades s'han identificat els següents compostos no descrits prèviament en mostres sedimentàries: 1-hidroxi-11-, 12-, 13- i 14-triacontanona, 1-hidroxi-16-dotriacontanona i 1-hidroxi-17-, 19- i 21-tetracontanona.

f) Entre els àcids carboxílics analitzats s'han trobat per primera vegada en una mostra geològica diversos compostos amb funcions oxigenades al mig de la cadena que representen una ampliació considerable, en quant a la varietat estructural dels àcids *n*-alcanoics prèviament descrits. Així, s'han identificat distribucions d'hidroxiàcids entre C25 i C35 amb el grup hidroxil situat en posicions centrals de la cadena hidrocarbonada. S'han trobat oxoàcids (p.e: àcid 17-oxotetratricosanoic), oxoàcids dicarboxílics (p.e: àcid 16-oxohentriacontandioic) i hidroxiàcids dicarboxílics (p.e: àcid 16-hidroxitetratricosanoic). Molt probablement tenen a un origen bacterià.

g) De manera temptativa s'ha identificat el 2,5,7,8-tetrametil-2-propilcromà (VII) que representa el primer compost cromànic de cadena curta (<C24) descrit en una mostra sedimentària.

h) De manera temptativa s'ha identificat la 3,9,13-trimetil-6-(1',5'-dimetilhexil)-2-tetradecanona (XVb), la primera cetona d'estructura isoprenoide "tipus T", descrita en mostres sedimentàries.

i) Entre els compostos isoprenoides d'aquest tipus d'estructura, també s'ha identificat de manera temptativa el 2-isobutil-4-[6'-(3',9',13'-trimetiltetradecil)]tiofè (XXXIXb).

j) Amb estructura isoprenoide regular s'han identificat de manera temptativa per primera vegada dos compostos de 15 i 35 àtoms de carboni amb un grup benzotiofènic: el 3,6-dimetil-2-(3-metilbutil)benzotiofè (XL) i el 3,6-dimetil-2-(3,7,11,15,19-pentameteicosanil)benzotiofè.

k) Entre els compostos d'estructura hopanoide s'ha identificat el 30-(2'-metilentiènil)hopà (LXXXI) i els seus homòlegs de 34 a 31 àtoms de carboni, el 30-[2'-(5'-metiltiènil)]hopà (LXXXII), el 30-(2'-metilentiolanil)hopà (LXXXIX) i el 30-[2'-(5'-metiltiolanil)]hopà (LXXX). Aquests compostos formen mescles d'esteroisòmers de configuració 17 β ,21 β (H), 17 α ,21 β (H) i 17 β ,21 α (H). Anteriorment a aquesta tesi només el 30-(2'-metilentiènil)-17 β ,21 β (H)-hopà havia estat descrit en mostres sedimentàries. Per tant la identificació de tots aquests compostos representa una notable extensió de la varietat estructural d'aquestes famílies d'organosofrats presents en mostres sedimentàries i una contribució al coneixement del seu cicle diagenètic.

Entre els productes d'aromatització diagenètica dels triterpenoides, les famílies de compostos noves més importants que s'han identificat comprenen:

l) Seco-C-des-A-triterpens. S'han trobat el 1-(2,6-dimetil-1-benzil)-2-(2,7,8-trimetil-1-naftil)età (CXVII), el 1-(2,6-dimetil-1-benzil)-2-(2,7-dimetil-1-naftil)età (CXVIII), 1-(2,6-dimetil-1-benzil)-2-(2,7,7-trimetil-1-(7,8-dihidronaftil)età (CXIX) i el 1-(2,6-dimetil-1-benzil)-2-[2,7,7-trimetil-1-(5,6,7,8-tetrahidronaftil)età (CXX) que representen els primers compostos d'aquest tipus d'estructura trobats en mostres sedimentàries.

o) Seco-C-triterpens. S'ha identificat el 1-(2,5,6-trimetil-1-naftil)-2-(2,7,8-trimetil-1-naftil)età (CVIII) i el 1-(2,5-dimetil-1-naftil)-2-(2,7,8-trimetil-1-naftil)età (CIX) que permeten completar la sèrie diagenètica d'aromatització dels seco-C-triterpens ja que fins ara els compostos amb estructura totalment aromatitzada eren desconeguts.

Pel que fa a les característiques deposicionals de les conques estudiades dins de la present tesi, l'anàlisi de la composició lipídica ha permès posar de manifest que:

a) La matèria orgànica sedimentària de les conques de Libros i Rubielos és dominada per aportats provinents de plantes superiors. Una part d'aquests ha experimentat un grau considerable de retreballament bacterià produït en medi òxic. En aquest sentit, les transicions entre sistemes deposicionals òxics i anòxics de la conca de Rubielos es reflecteixen en diversos canvis de la composició lipídica sedimentària que permeten establir alguns índexs d'oxicitat: $17(21)\text{-hopè}/17\beta,21\beta\text{-hopà}$; $2\text{-colestè}/\text{octacosà}$ i $6,10,14\text{-trimetil-2-pentadecanona}/2\text{-pentacosanona}$.

b) Els aportats provinents de plantes superiors també són predominants a la conca de Campins encara que aquesta es distingeix de les anteriors per l'elevat caràcter reductor del medi deposicional. En aquestes condicions, els processos de sulfatorreducció foren molt importants i es produí una quantitat notable de compostos organosofrats que altrament només són generats en sistemes hipersalins.

c) Els hidrocarburs sofrats també són abundants a la conca de Ribesalbes, que d'acord amb la seva composició lipídica correspon a un ambient deposicional hipersalí. En aquesta conca predominen els aportats algals, que es reflecteixen en una gran proporció de compostos isoprenoides altament ramificats de tipus T. Els aportats bacterians també es troben en gran abundància i són representats per quantitats elevades de compostos d'estructura hopanoide.

d) A la conca de la Cerdanya predominen aportats terrestres i bacterians. Els primers es troben en quantitats importants a gairebé tota la seqüència sedimentària, però els segons esdevenen majoritaris a les fàcies on s'han generat fosforites. La gradació d'oxicitat dins d'aquesta conca és en gran part relacionada amb la distància al centre deposicional de l'antic llac. Així les mostres situades en fàcies laterals corresponen a condicions deposicionals òxiques i els sediments localitzats en fàcies centrals presenten uns ambients anòxics. No obstant això, tant a uns com a altres es troben seqüències de formació de fosforita.

Finalment, l'estudi de la composició lipídica en aquestes conques lacustres terciàries també ha permès aprofundir en el coneixement d'alguns dels processos biogeoquímics que es produeixen durant la diagènesi. Mereixen especial atenció els resultats assolits en relació a:

- a) les vies diagenètiques d'aromatització de compostos triterpenoides,
- b) els processos de formació de compostos organosofrats i
- c) les condicions deposicionals per a la generació de fosforites.

a) Els compostos triterpenoides identificats a la conca de Rubielos han permès establir algunes etapes intermèdies encara desconegudes entre els processos d'aromatització dels lípids sedimentaris. Així, la identificació del 2,2,4a,6a,9-pentametil-1,2,3,4,4a,5,6,6a,7,8,13,14,14a,14b-tetradecahidropicè (XCIII) i del 2,2,9-trimetil-1,2,3,4,5,6-hexahidropicè (XCVIII) han posat de manifest que durant la formació d'estructures parcialment aromatitzades, es pot produir la pèrdua de grups metil angulars, simultàniament a la formació de dobles enllaços conjugats. En un estadi posterior de maduració, l'aromatització del cicle desmetilat continua mitjançant la formació de més insaturacions. Aquesta via representa un camí alternatiu a la pèrdua de metil quan s'acompleix l'aromatització total d'un anell addicional, que és la via diagenètica descrita fins ara.

b) La presència abundant de compostos organosofrats a les conques de Ribesalbes i Campins, i a les zones més reductores de la conca de la Cerdanya, posa de manifest que malgrat anteriors presumpcions, la formació d'aquests compostos no va ineludiblement associada a sistemes deposicionals hipersalins. Elevada anòxia, absència de ferro o d'altres metalls pesants i disponibilitat de sulfats, semblen les tres condicions principals per a la generació de compostos organosofrats. En aquest sentit, a la conca de Libros aquests compostos no es troben. El sistema deposicional òxic, no ho permeté malgrat la gran disponibilitat de sulfats. Els compostos organosofrats en general es formen per combinació d'espècies reduïdes de sofre amb lípids bacterians. No s'han observat diferències significatives que permetin distingir de manera selectiva els compostos organosofrats generats en sistemes hipersalins.

c) Els dipòsits fosfàtics de la Cerdanya foren generats en condicions subòxiques, tal com ho posa de manifest la proporció de carboni orgànic que correspon a les capes de fosforita (COT entre 0.5 i 1.3%). En aquest sentit, en seqüències deposicionals òxiques (COT 0.3-0.4%) la generació de fosfats s'observa en períodes intermedis de menor grau d'oxidació (COT 0.6-0.8%). En canvi, en seqüències anòxiques (COT 2.5-3.5%), la formació de fosfats es produeix en transicions d'anòxia a subòxia (COT 1.3%).

Independentment del fet que els fosfats s'hagin incorporat a aquests sediments en associació amb:

- a) la matèria orgànica detritica d'origen terrestre,
- b) els residus sedimentaris de diatomees,

la importància de l'activitat microbiana en relació amb la fosfatogènesi és ben palesa. Aquests aportats microbians són especialment significatius a les seqüències òxiques, on els increments de carboni orgànic i fosfat es reflecteixen en augments molt importants de la proporció d'hopans. Aquests resultats indiquen que la generació de fosfat és un fenomen molt més dependent de les condicions ambientals de deposició i dels processos diagenètics, que dels aportats específics de matèria orgànica o de la composició mineralògica del sediment.

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