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From mass to structure: Thermogenic organic matter dissolved in the ocean

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Formation and decay of thermogenic organic matter are important processes in the geological carbon cycle, but little is known about the origin and fate of combustion-derived and petrogenic polyaromatic hydrocarbons (PAHs) in the ocean. We explored the molecular structure of marine dissolved organic matter (DOM) on a thermogenic signature in different water masses of the Southern Ocean by ultrahigh-resolution Fourier transform ion cyclotron resonance mass spectrometry (FT-ICR-MS). For structural identification of aromatic sub-structures in natural organic matter we propose a general aromaticity index (AI) and two threshold values as unequivocal criteria for the existence of either aromatic (AI $>$ 0.5) or condensed aromatic structures (AI $>=$ 0.67). AI can be calculated from molecular formulae which are derived from exact molecular masses of naturally occurring compounds containing C, H, O, N, S and P. More than 200 different PAHs were identified, most of them consisting of seven condensed rings with varying numbers of carboxyl, hydroxyl, and aliphatic functional groups. These unambiguously thermogenic compounds were homogenously distributed in the deep sea, but depleted at sea surface. Turnover rate and surface depletion point toward a primarily petrogenic source, possibly deep-sea hydrothermal vents, although structural information alone is not sufficient to distinguish between petrogenic and pyrogenic origin of PAHs in DOM. We estimate that $>$2.4% of DOM are thermogenic compounds, and their turnover is $>1.2 \cdot 10^{12}$ mol C per year, significantly impacting global biogeochemical cycles.