



Direct Molecular-scale Insight into Soluble Organic Matter from the Murchison and Aguas Zarcas Meteorites Enabled by 21T FT-ICR MS and Single-molecule HR-AFM Imaging

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Abstract

Organic material in the Murchison and Aguas Zarcas meteorites preserves molecular records of early solar system chemistry and potential precursors to life. Here, we present an ultra-wide-mass-range molecular analysis of their soluble organic matter (SOM), complemented by high-resolution structural imaging, revealing large and complex soluble hydrocarbon molecules, including Fe-bearing species extending to carbon numbers above C₃₅. We employ polarity-graded sequential solvent extraction combined with 21 Tesla Fourier-transform ion cyclotron resonance mass spectrometry (21T FT-ICR MS). More polar solvents preferentially extract heteroatom-rich, more aliphatic compounds, whereas nonpolar solvents enrich lower-heteroatom, more aromatic species. Over 18,000 molecular formulas were assigned per extract, with mass errors below 80 ppb, sorted into CHO, CHNO, CHNOS, and CHOMg classes. Carbon numbers range from C₁₀–C₇₀, with oxygen contents up to O₂₀. Differences in Fe-associated molecular populations between Murchison and Aguas Zarcas indicate distinct metal-organic interaction regimes. Negative-ion electrospray ionization (ESI) highlights abundant oxygenated species, with the acidic O₂ class comprising up to 20% of assigned formulas. In assisted positive-ion ESI, N_yO_x species dominate, and extensive sodium adduct formation is observed. Organomagnesium compounds are detected primarily in methanol extracts for Murchison and in chloroform extracts for Aguas Zarcas. Aromaticity increases with decreasing solvent polarity. High-resolution noncontact atomic force microscopy (HR-AFM) provides structural images of representative molecules, spanning saturated chains to condensed aromatic cores. The achieved resolution supports detailed structural interpretation and future molecular-level structure determination.

Unified Astronomy Thesaurus concepts: Astrochemistry (75); Carbonaceous grains (201); Cosmochemistry (331); Cosmic abundances (315); Interstellar dust (836); Meteorite composition (1037); Polycyclic aromatic hydrocarbons (1280); Meteorites (1038); Mass spectrometry (2094); Experimental techniques (2078)

1. Introduction

Meteorites provide a glimpse into the past of the solar system by preserving snapshots of the Universe for millions to billions of years (M. A. Sephton 2005). Organic material forms over this time, creating common molecules seen on Earth such as carboxylic acids, aromatic and aliphatic hydrocarbons, amino acids, and components relevant to carbohydrates, as well as DNA and RNA chemistry (M. A. Sephton 2005; S. Pizzarello 2006; S. Pizzarello et al. 2006; S. Pizzarello & E. Shock 2010; Y. Oba et al. 2022; Y. Furukawa et al. 2026; T. Koga et al. 2026). Organic material formed in such a way is often revealed as ultracomplex mixtures, similar to fossil fuels and natural dissolved organic matter seen on Earth.

Recent sample-return missions have changed the study of primitive solar system organic matter by providing carbonaceous asteroid material with minimal exposure to Earth's atmosphere and biosphere. Hayabusa2 returned samples from asteroid Ryugu that were handled under vacuum or purified nitrogen and shown to preserve physical and spectral

properties representative of the asteroid surface. These studies established that Ryugu is closely related to CI-like carbonaceous material, with a highly porous and fragile character, and molecular analyses of its soluble organic matter (SOM) revealed extensive organic diversity, including approximately 20,000 molecular species. More recently, OSIRIS-REx returned approximately 120 g of pristine carbonaceous regolith from Bennu, confirming the presence of hydrated phyllosilicates, magnetite, carbonates, organic compounds, sulfides, presolar grains, and phosphate phases. These returned samples provide important ground truth for primitive asteroid chemistry and create a necessary reference point for interpreting organic matter in meteorites that have experienced terrestrial exposure (L. D. Tunney et al. 2022). Meteorites such as Murchison and Aguas Zarcas remain essential reference materials because they are available in larger quantities and have been extensively studied across both soluble and insoluble organic fractions. At the same time, unlike returned asteroid samples, meteorites recovered on Earth can be affected by atmospheric exposure, humidity, handling, storage conditions, and other forms of terrestrial alteration. These differences do not reduce their value, but they do require careful interpretation when comparing meteorite organic matter with pristine returned samples. In this context, high-resolution molecular characterization of meteorite SOM remains important, particularly when the analytical workflow

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is designed to increase molecular coverage and to evaluate how the extraction solvent and ionization mode influence the observed composition (T. Yada et al. 2021; S. Tachibana et al. 2022; Y. Oba et al. 2023; D. S. Lauretta et al. 2024).

One of the most primitive classifications of meteorites is carbonaceous chondrites (CCs), which reveal the highest quantity of organic matter (up to 6% by weight) as both insoluble and soluble material (V. K. Pearson et al. 2006). However, the organic material varies in the subclassification of CCs, with eight subcategories (D. W. Mittlefehldt 2008). Several CC groups, including CI, CM, CR, and some C-ungrouped meteorites, contain significant organic matter. Here, we focus on CM chondrites, including Murchison and Aguas Zarcas, which are important reference materials for studies of meteoritic SOM and insoluble organic matter (IOM). This meteorite has been extensively studied in both insoluble and soluble organics with a variety of analytical techniques. IOM has been characterized by a range of molecular, isotopic, and spectroscopic approaches, including destructive methods after mineral dissolution, such as nuclear magnetic resonance (NMR), pyrolysis GC-MS, elemental/isotopic analysis, and GC-IRMS, as well as nondestructive or minimally destructive methods such as Raman spectroscopy, FTIR, and XANES (S. Pizzarello 2006; H. Busemann et al. 2007; C. Alexander et al. 2017; G. Danger et al. 2020). SOM is commonly analyzed after sample homogenization followed by extraction with water, organic solvents, or acidified solvents, often assisted by heat or sonication, depending on the compound classes of interest (P. Schmitt-Kopplin et al. 2010; A. A. Monroe & S. Pizzarello 2011; D. N. Simkus et al. 2019). These workflows may also include purification, derivatization, liquid chromatography, and mass spectrometry analysis. Different extraction solvents and conditions access different fractions of the soluble organic mixture rather than representing entirely separate analytical categories.

Techniques of analysis vary based on whether the study is targeted or untargeted. Targeted reports have focused on single or multiple classes of compounds such as carboxylic acids, amino acids, hydrocarbons, N-heterocycles, nucleobases, and organosulfur compounds (S. Pizzarello et al. 2006; A. A. Monroe & S. Pizzarello 2011; M. P. Callahan et al. 2014). These workflows are typically optimized for selected compound classes and often rely on class-specific extraction or derivatization, chromatographic separation, authentic standards, retention-time comparison, and/or tandem MS for compound-level identification and quantification. Such approaches provide detailed structural and concentration information for selected molecular families, but they do not capture the full formula-level diversity of the soluble organic mixture. Methods such as GC-MS, LC-MS, GC-IRMS, and GC $\times$ GC-MS can identify and quantify individual molecular structures and, in some cases, provide compound-specific isotope information. In contrast, nontargeted ultra-high-resolution Fourier-transform ion cyclotron resonance mass spectrometry (FT-ICR MS) provides broader formula-level coverage of ultracomplex soluble organic mixtures, revealing tens of thousands of distinct compositions, but molecular formulas alone do not define structural isomers or compound-specific abundances. These approaches are therefore complementary and provide different levels of molecular-level information.

The present study follows the latter, nontargeted approach and does not attempt compound-specific identification of amino acids, nucleobases, or other small, targeted prebiotic

molecules. Those compounds require dedicated extraction and separation-based workflows, including chromatographic retention-time comparison, authentic standards, and, in many cases, derivatization and/or tandem MS. In addition, the canonical nucleobases fall below approximately 155 Da, which is below the effective broadband mass range used here for 21 Tesla (21T) FT-ICR MS analysis. Therefore, the molecular assignments reported in this work should be interpreted at the formula and heteroatom-class levels rather than as structural identifications of specific amino acids or nucleobases.

Several reports have focused on molecular-level characterization of meteorites and have been previously performed on both Murchison and Aguas Zarcas. Those studies used the mass analyzer with the highest mass accuracy and resolving power: FT-ICR MS. For instance, P. Schmitt-Kopplin et al. (2010) focused on the identification of solvents for optimal extraction of Murchison SOM based on various degrees of solubility and dipolar moments, in order to achieve comprehensive analysis by negative-ion (−) electrospray ionization (ESI) coupled to 12 Tesla (12T) FT-ICR MS. Another report, by J. Hertzog et al. (2019), focused on investigating the ionization mode that could provide the highest number of molecular formula assignments. In both cases, it was determined that extraction with methanol followed by (−) ESI FT-ICR MS provided the best results, with ~10,000–11,000 assigned formulas. Such methodology has been used for several studies, such as those reported by N. Hertkorn et al. (2015) and A. Ruf et al. (2017), which revealed more than 5600 formulas and similar compositional trends.

The Ryugu molecular atlas reported by P. Schmitt-Kopplin et al. (2023a) further demonstrated that sequential solvent extraction combined with complementary ionization methods and FT-ICR MS can reveal extensive formula-level diversity in returned asteroid SOM, while also linking solvent- and ionization-dependent molecular distributions to parent-body aqueous alteration and mineral–organic processing.

Subsequent studies on extraterrestrial organic matter include the reports by I. Kerraouch et al. (2022) for the examination of the Aguas Zarcas meteorite and P. Schmitt-Kopplin et al. (2023b) for the analysis of Martian meteorites. These two studies revealed mass spectra with ~5,000 and ~10,000 molecular formula assignments, respectively; more recently, FT-ICR MS was applied to methanol extracts of pristine Bennu material returned by OSIRIS-REx, revealing approximately 16,000 molecular formulas containing C, H, N, O, S, and Mg. That study showed that Bennu SOM is particularly enriched in nitrogen-containing species, in contrast to the sulfur-rich chemistry reported for Ryugu, and further demonstrated the value of ultra-high-resolution mass spectrometry (UHR-MS) for comparing organic inventories across returned asteroid samples and carbonaceous meteorites (M. A. Sephton 2005; S. Pizzarello 2006; S. Pizzarello et al. 2006; S. Pizzarello & E. Shock 2010; T. Yada et al. 2021; Y. Oba et al. 2022, 2023; S. Tachibana et al. 2022; H. Naraoka et al. 2023b; D. S. Lauretta et al. 2024; T. Koga et al. 2024, 2026; C. J. Yen et al. 2024; D. P. Glavin et al. 2025; Y. Furukawa et al. 2026).

Several analyses have also been performed on the IOM of the Paris and Aguas Zarcas meteorites with Laser Desorption Ionization (LDI) 12T FT-ICR MS. These reports identified ~14,000 formulas in the Paris meteorite and ~8700 formulas for Aguas Zarcas (G. Danger et al. 2020; B. Laurent et al.

2022). Similar compound classes, e.g., O_x , N_yO_x , S_zO_x , etc., were observed in comparison to SOM, but with much higher carbon content and aromaticity. Fullerenes were detected by LDI MS at high m/z (between m/z 600 and 1200) but increased in abundance when under high laser power (G. Danger et al. 2020; B. Laurent et al. 2022). Importantly, fullerenes were not frequently found in broadband ESI-MS analysis, and it should be considered that high-powered laser irradiation of carbonaceous materials can produce fullerene-like structures, as shown by T. M. Pereira et al. (2014) with asphaltenes as models (S. R. Wilson & Y. Wu 1993a, 1993b; G. Khairallah & J. Peel 1998; V. G. Santos et al. 2016).

The National High Magnetic Field Laboratory provides a unique advantage in the analysis of ultracomplex mixtures as it is the home of the world's highest-field and highest-resolving custom-built 21T FT-ICR MS (hereafter 21T-MS; C. L. Hendrickson et al. 2015). This instrument has been used extensively for the characterization of ultracomplex terrestrial samples since installation in 2015 (C. L. Hendrickson et al. 2015; D. F. Smith et al. 2018). Extensive studies have focused on fossil fuels and environmental samples, some of the most complex terrestrial mixtures that can be found (D. F. Smith et al. 2018). For such mixtures, mass spectral characterization by 21T-MS has revealed up to over 49,000 assignments for a heavy distillate cut from fossil fuel and up to over 24,000 assignments for Suwannee River fulvic acid (SRFA; D. F. Smith et al. 2018; W. Bahureksa et al. 2022). The 21T-MS routinely resolves peaks whose masses differ by a few milliDalton (mDa). For instance, the 1.1 mDa mass split corresponds to a compositional difference of $H_3S_1^{13}C$ and C_4 . Another mass split reported in fossil fuels, even at high m/z , is 0.53 mDa (less than the mass of an electron), which originates from peaks that differ in C_5N_1 and $H_8S_1^{34}S_1$ content (D. F. Smith et al. 2018). Fossil fuel applications have included the analysis of asphaltenes, an extraordinarily complex mixture that has shown more than 500 peaks at one nominal mass (m/z 627) (M. L. Chacón-Patiño et al. 2017, 2021). Environmental applications have focused on the analysis of field samples from oil spills, natural dissolved organic matter, and per- and polyfluoroalkyl substances (PFASs), all of which take advantage of the unique resolving power provided by the 21T-MS. Terrestrial dissolved organic material from SRFA has proven to be just as complex as petroleum samples, with up to 24,000 peaks and mass splits of only 0.43 mDa ($[C_{23}H_{14}O_{17}C_1]^{2-}$ versus $[S_1]^{1-}$) (W. Bahureksa et al. 2022).

High-resolution noncontact atomic force microscopy (HR-AFM) can complement the analysis of meteorite organic matter and other extraterrestrial samples by resolving the structures of a few selected individual molecules (Figure C1) within complex molecular populations, as first shown for Murchison SOM by K. Kaiser et al. (2022). Using CO-functionalized tips (B. Lassagne et al. 2009), HR-AFM has been applied to a wide range of complex molecular systems, including soot, marine dissolved organic matter, asphaltenes, and astrochemical analogs (L. Gross et al. 2010; K. Ø. Hanssen et al. 2012; D. G. de Oteyza et al. 2013; B. Schuler et al. 2015, 2017a; S. Fatayer et al. 2018; M. Commodo et al. 2019; F. Schulz et al. 2021). Most recently, K. Iwata et al. (2026) reported HR-AFM imaging of SOM from asteroid Ryugu, revealing large, nonplanar polycyclic aromatic hydrocarbon (PAH)-like molecules containing five- to eight-membered

rings. These results demonstrate that single-molecule imaging can uncover aromatic structures significantly larger and more diverse than those identified by GC/MS or FT-ICR MS, with estimated core sizes ranging from a few fused rings to over 100 rings. This helps bridge the gap between relatively small PAHs identified in returned samples and the larger populations inferred from astronomical observations of the interstellar medium.

Beyond structural discovery, HR-AFM provides valuable ground-truth validation for techniques such as FT-ICR mass spectrometry in complex heterogeneous mixtures (B. Schuler et al. 2017a). It enables direct, isomer-specific identification of proton-poor compounds that are difficult to resolve by mass spectrometry or NMR (L. Gross et al. 2010; K. Ø. Hanssen et al. 2012), and can even detect rare species due to its single-molecule sensitivity. However, despite its ability to resolve aromatic carbon skeletons with submolecular precision, HR-AFM alone has limitations: The identification of heteroatoms (e.g., N, S) and peripheral functional groups (e.g., $-CH_3$, $-OH$, $-COOH$, $-NHR$, etc.)—especially in nonplanar molecules—remains challenging without reference compounds or advanced image simulations. These include approaches such as the full density-based model (FDBM; M. Ellner et al. 2017, 2019), which enables ab-initio-level simulation of AFM contrast from candidate molecular structures derived from theoretical models. Interpretation therefore still relies in part on expert judgment.

Recent efforts toward automation primarily focus on the imaging process itself, enabling efficient and reproducible acquisition of large numbers of molecular images (S. Arias et al. 2023). In contrast, translating AFM images into molecular structures remains a major bottleneck and is still largely manual. While machine learning approaches show promise for assisting AFM image-to-structure interpretation, this area is still under active development.

Here, we use the 21T-MS to characterize the nonterrestrial SOM extracted from the Murchison and Aguas Zarcas meteorites, fully leveraging its analytical advantages. Complementing this approach, HR-AFM is employed to directly image molecular structures of species extracted from Murchison. Previous FT-MS studies of meteorite and returned asteroid SOM have shown the value of solvent selection, sequential extraction, and complementary ionization methods for expanding molecular coverage (P. Schmitt-Kopplin et al. 2010, 2023a). Building on this foundation, we use a polarity-graded sequential extraction, followed by concentration-normalized positive- and negative-ion (+/−) ESI FT-ICR MS with ionization modifiers, to expand formula-level coverage of Murchison and Aguas Zarcas SOM across chemically distinct solvent fractions.

Here, we use a polarity-graded sequential extraction, followed by concentration-normalized positive- and negative-ion ESI FT-ICR MS with ionization modifiers, to expand formula-level coverage of SOM across chemically distinct solvent fractions. Although sequential and polarity-based extractions have been used in targeted studies to recover specific compound classes, the present workflow combines solvent polarity fractionation, both ESI polarities, and 21 T resolving power to map a broader compositional space of meteorite SOM. This method yields tens of thousands of unique compounds for each meteorite across a range of compositional classes. Both meteorites demonstrate increased

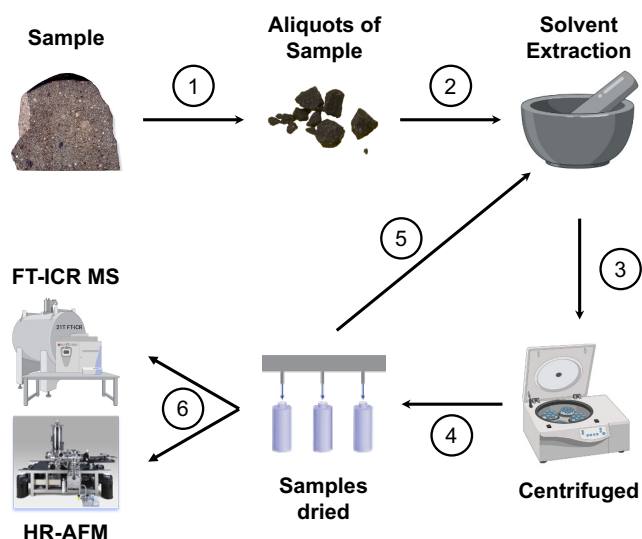


Figure 1. Experimental schematic for sequential extraction of soluble organic material from meteorites for analysis by FT-ICR MS and HR-AFM.

molecular formula assignments with the shift toward a nonpolar solvent, with Murchison assigning the most formulas in chloroform and Aguas Zarcas in toluene. This was reflected in van Krevelen plots, with an increase in aromaticity in the nonpolar solvents. Increased aromaticity was confirmed with AFM images showing an increase in aromatic structures observed from chloroform to an acetone extract. Organometallic species such as oxygenated magnesium and iron compounds were also observed. The introduction of these metals increased the complexity of these samples, which resulted in an observed mass split of 0.36 mDa, highlighting the necessity of the resolving power provided by the 21T-MS.

2. Experimental Methods

2.1. Materials

A Murchison meteorite fragment (ME 2644 #23.6) was obtained from the Robert A. Pritzker Center for Meteoritics and Polar Studies at the Chicago Field Museum. An Aguas Zarcas fragment (ASU #2121 from AZMF10) was obtained from the Buseck Center for Meteorite Studies at Arizona State University. High-performance liquid chromatography (HPLC)-grade chloroform (CHCl_3 ; J.T. Baker) and toluene (Tol; J.T. Baker) were obtained and used as received. Liquid chromatography–mass spectrometry-grade ethanol (EtOH; LiChrosolv by Supelco) and methanol (MeOH; Burdick & Jackson) were obtained and used as received. Formic acid (FA; Pierce) and tetramethylammonium hydroxide (TMAH, 25 w% in MeOH; Acros Organics) were used to aid in the ionization process.

2.2. Extraction of Organic Material

A sequential solvent extraction was performed following the experiment outline in Figure 1. Entire fragments of approximately 2 g are homogenized into a near powder state (greater than $0.45 \mu\text{m}$) (1) and aliquots of 30–40 mg are weighed out. The aliquots are then washed with solvent (2) prior to extraction in an agate mortar and pestle with 1 mL of solvent. All the material is centrifuged (3), after which the supernatant

is collected. Both the supernatant and remaining material are dried under a gentle stream of nitrogen gas.

The drying step was used to remove extraction solvent, minimize solvent carryover between sequential extractions, and allow reconstitution of each extract at a controlled concentration before FT-ICR MS analysis. This workflow was designed for nonvolatile and semivolatile SOM detectable by broadband FT-ICR MS and HR-AFM, and was not intended to preserve or quantify highly volatile organic compounds.

The remaining solid material is then washed and extracted with the next solvent (5) to extract additional organics with decreasing dipolar moment. The solvents used were methanol, ethanol, chloroform, and toluene. All the supernatants were reconstituted in the extraction solvent (6) prior to analysis. An additional extraction with all remaining Murchison material was performed with acetone, as asphaltenes demonstrated the selective extraction of planar PAH compounds with this solvent (M. L. Chacón-Patiño et al. 2020).

2.3. Molecular Characterization by FT-ICR MS

All samples were concentration-normalized to $250 \mu\text{g mL}^{-1}$ in 50:50 methanol/extraction solvent. To aid in ionization, formic acid (+ESI) and TMAH (−ESI) were spiked in at 1% and 0.0125%, respectively. Samples were directly infused at $0.5 \mu\text{L min}^{-1}$ for positive-ion (+) and negative-ion (−) ESI analysis by a custom-built hybrid Velos Pro 21T-MS (C. L. Hendrickson et al. 2015).

Microelectrospray ionization was selected because the meteorite extracts were sample limited and could be analyzed at $0.5 \mu\text{L min}^{-1}$. Although APCI and APPI are better suited for nonpolar aromatic hydrocarbons, the available Ion Max APCI/APPI source required substantially higher flow rates, approximately $50 \mu\text{L min}^{-1}$, which was not compatible with the limited extract volumes available for this study.

Approximately 2.0×10^6 ion charges were externally accumulated with a Velos Pro dual linear ion trap prior to transfer to a dynamically harmonized ICR cell operated at a 6 V trapping potential. Due to sample-yield limitations, the acetone extraction was only analyzed with (−) ESI. When necessary, a m/z -specific ion-depletion waveform was utilized to minimize chemical noise peaks and maximize the analyte peaks in the accumulation process (C. R. Weisbrod et al. 2024). The advantages of data collection with ion depletion, or gas-phase depletion of contaminant ions, have been highlighted in a previous work (D. M. Nisson et al. 2023). Figure A1 displays an example of why this is necessary with an ethanol extract of Murchison. Without the ion depletion, only 16,105 peaks were found in the spectrum. Depletion was activated on the five most abundant chemical contaminant peaks found in methanol for the (−) ESI analysis, highlighted in the figure. Use of the depletion waveform increased the number of peaks to 23,104 peaks for the same sample. A Predator data station coadded 400 time-domain transients of 3.1 s for each sample (D. F. Smith et al. 2018). Data collection, Fourier transformation, rephasing, external calibration, molecular formula assignment, and data visualization were performed with custom Predator and PetroOrg[®] software (F. Xian et al. 2010; G. T. Blakney et al. 2011; Y. E. Corilo 2014). Theoretical isotopic fine structure was calculated with IsoPro 3.1.

2.4. LT-STM/AFM System

AFM measurements were performed using a CreaTec-based low-temperature scanning tunneling microscopy (LT-STM) system (CreaTec Fischer & Co. GmbH, Berlin, Germany) custom-upgraded with HR-AFM capability and operated with the open-source GXSM control software (P. Zahl et al. 2003, 2010; P. Zahl & T. Wagner 2026). The ultrahigh vacuum (UHV) system had a base pressure of $<9 \times 10^{-11}$ mbar. The qPlus sensor used for this work featured a resonance frequency of 30,210 Hz, a spring constant of $\sim 1800 \text{ N m}^{-1}$, and a quality factor of the order of 10,000 when far from the surface (F. J. Giessibl 2019, 1998). A 25 μm -diameter platinum-iridium wire, cut and sharpened by focused ion beam milling, was attached to the qPlus sensor and served as the tip. Before being mounted on the SPM scanner, the tip was cleaned by Ar⁺ sputtering from three directions under UHV conditions. Final shaping of the metal tip apex was performed in situ via controlled nanoindentations into the metal single crystal surface, accompanied by bias pulsing.

2.5. Sample Preparation for STM/AFM

An ultraclean metal single crystal surface was prepared first. We experimented with Ag(111), Au(111), and Cu(111) single crystal substrates, as they provide different advantages and disadvantages for stabilizing molecules of various kinds. Those were refreshed (previously cleaned, but containing few molecules from previous work) by four cycles of Ar⁺ sputtering followed by annealing; in each cycle, sputtering was carried out for 20 minutes with an ion current of 3.5 μA at 1 kV, followed by annealing up to 550°C for another 10–15 minutes. The substrate was then inserted into the microscope and cooled down to 5 K, and confirmed to be clean by STM.

2.6. Molecule Source Preparation

A highly doped silicon wafer strip ($\sim 4 \text{ mm} \times 15 \text{ mm}$) was used as the sample material carrier. It was cleaned in UHV by direct-current heating to temperatures well above the typical peak flash temperature. A cleaned titanium tweezer tip was used to transfer a “hint of stardust” from the extracted sample material from a vial onto this carrier simply via touching both. This procedure transferred an ideal amount of sample material to adhere near the center of the wafer strip, after which it was loaded into the UHV system.

2.7. Molecule Deposition

A low surface coverage ($\ll 1\%$ ML) of individual sample molecules was prepared by thermal evaporation via flash heating from this silicon wafer strip onto the line-of-sight metal crystal surface inside the microscope’s cryoshield from a distance of about 15 cm (Y. Zhang et al. 2018). The sample temperature rises briefly to about $\sim 10 \text{ K}$ while the cryo’s door is opened for this process.

2.8. CO Tip Functionalization

The prepared substrate was briefly exposed to a low dose (5 s at $\sim 2 \times 10^{-8}$ mbar) of CO gas to add a “small number” of CO molecules for tip functionalization to the mix. Scanning on surface areas with potential CO molecules while avoiding other molecules at low bias ($\leq 100 \text{ mV}$ around 10 pA) at

chance creates a CO-terminated tip. This process can be time-consuming as it depends on the chance of locating a suitably “low-coordinated” CO molecule. Many repetitions of tip reforming via controlled tip-surface contact and CO pickup attempts are usually required.

2.9. HR-AFM Imaging

After successful preparation and confirmation of a CO-terminated tip, the system is ready for HR-AFM imaging. A large-scale overview scan (as shown in Figure C1) provides an initial assessment of the molecular shapes and enables selection of promising candidates for high-resolution imaging. Starting from a safe tip–molecule separation, a series of images is acquired while progressively decreasing the tip–molecule distance until satisfying detail is revealed. For more reactive or radical species, there is a high probability of losing the CO termination during imaging at very small tip–molecule distances, as required for ideal imaging. Similar issues arise when nonplanar molecules with significant out-of-plane side groups, for example CH₃ or single-bonded and out-of-plane-twisted phenyl groups, are encountered.

3. Results and Discussion

3.1. FT-ICR MS Analysis

An initial extraction method from broken-off pieces of a larger Murchison fragment was used to isolate organic species for 21T-MS analysis. The results revealed over 20,000 peaks with a signal magnitude higher than 6 times the average baseline noise level (i.e., $>6\sigma$). To test the reproducibility, two replicates were performed. Slight changes were made between the initial method and subsequent extractions as experiments between these displayed inconclusive results in terms of complexity observed. The spectra contained fewer than 8000 peaks, including electronic and chemical noise. Therefore, the entire experiment design was revisited. Homogenization of the entire fragment proved to be the solution to this issue, which allowed for similar compositional trends to be observed for replicates 1 and 2 of the final reported method, presented in Figure A1. Sample complexity is displayed across the entire mass range, shown in Figure 2, for the methanol extract of the three Murchison and one Aguas Zarcas extraction series. From low to high m/z , peak locations shift toward higher mass defect, which indicates the presence of molecules that are more aliphatic in nature. Tens of peaks are present at each nominal mass from m/z 175 to m/z 1000 for Murchison, which yielded tens of thousands of peaks detected over the entire mass range in each extract, as detected in both positive- and negative-ion modes. Aguas Zarcas features similar molecular complexity but with a narrower mass range than Murchison, at m/z 175 to 750; however, such samples reveal more peaks per nominal mass, which results in more detected compositions in each mass spectrum. The most abundant peak in each 15 mDa section demonstrates the ultralow error in molecular formula assignment that is present across the entire mass range. The number of detected peaks, assigned molecular formulas, and relative abundances assigned in each spectrum are presented in Table 1 for each sample. The last column contains the total number of unique assignments found between all four extracts.

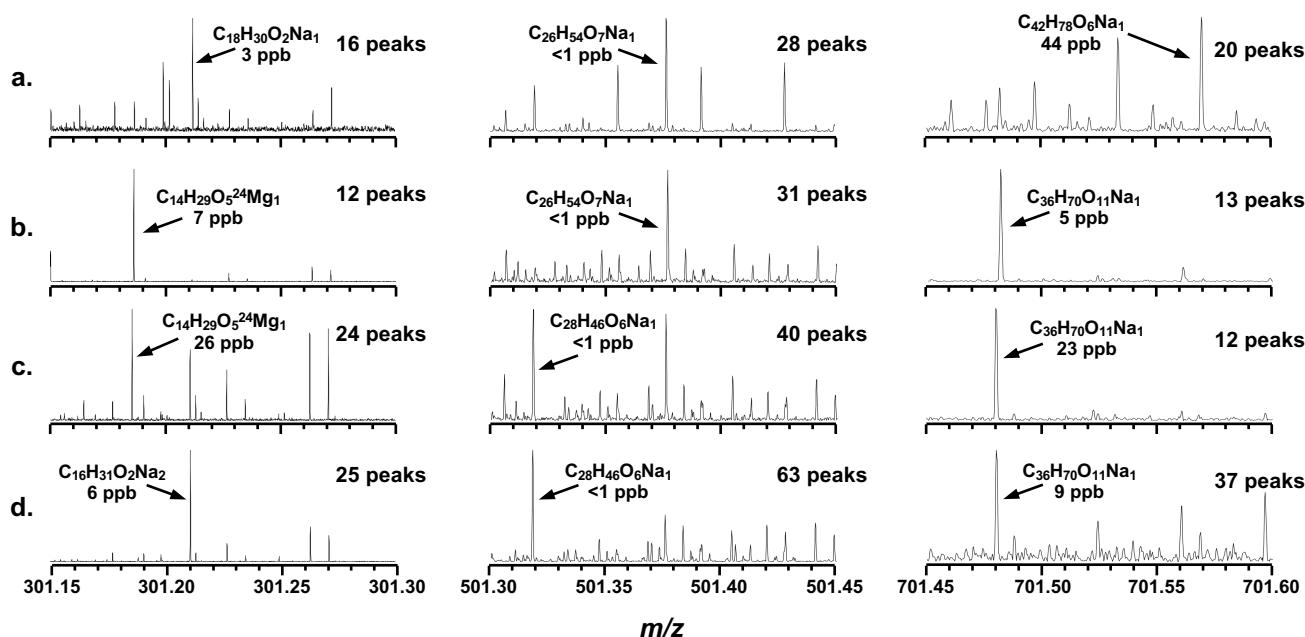


Figure 2. Zoomed sections of 15 mDa at m/z 301, 501, and 701 from the (+) ESI 21T-MS analysis of the methanol extractions of Murchison (a) IM, (b) R1, and (c) R2, and Aguas Zarcas (d). The number of peaks present in each 15 mDa section is shown for each segment, as well as the assignment for the most abundant peak with the corresponding error.

Table 1

The Number of Assignments Found in Each Extraction Solvent from the Murchison Initial Method, the Murchison Two Replicates, and Aguas Zarcas

(+) ESI	Methanol	Ethanol	Chloroform	Toluene	Unique
Murchison (I)	29,000 / 23,388 / 82.1%	38,761 / 31,917 / 82.8%	43,792 / 36,543 / 75.3%	35,376 / 28,607 / 73.1%	70,671
Murchison (R1)	31,508 / 19,860 / 71.0%	28,388 / 21,442 / 81.5%	44,644 / 35,289 / 72.9%	37,658 / 31,146 / 83.4%	66,187
Murchison (R2)	28,169 / 21,124 / 78.2%	38,371 / 27,316 / 76.6%	48,065 / 38,084 / 71.4%	38,358 / 29,643 / 81.2%	62,027
Aguas Zarcas	43,754 / 36,041 / 88.5%	48,145 / 36,756 / 81.7%	56,450 / 44,711 / 80.8%	59,619 / 48,444 / 86.3%	91,983
(-) ESI	Methanol	Ethanol	Chloroform	Toluene	Unique
Murchison (I)	27,874 / 18,286 / 72.1%	25,604 / 19,107 / 66.9%	29,329 / 23,073 / 81.0%	28,354 / 23,085 / 85.1%	54,954
Murchison (R1)	49,257 / 35,054 / 63.9%	45,929 / 35,009 / 76.3%	52,197 / 42,128 / 80.8%	40,135 / 30,298 / 83.4%	92,878
Murchison (R2)	34,077 / 23,275 / 63.0%	36,502 / 29,196 / 82.5%	41,022 / 30,979 / 78.4%	30,699 / 22,475 / 70.5%	57,483
Aguas Zarcas	47,553 / 39,970 / 79.0%	49,786 / 41,733 / 84.4%	54,014 / 40,175 / 79.9%	42,475 / 32,532 / 79.2%	82,783

Note. Included are the number of unique assignments for each sample, as well as the number of detected peaks, assignments, and percent relative abundances assigned for the first four extracts from (+) and (−) ESI. The total number of unique formula assignments for each extraction solvent is found in the last column.

3.2. Molecular Formula Assignments

Peaks were assigned a unique molecular formula based on ultrahigh mass accuracy, with a mass error window of ± 0.15 ppm for all datasets. Assignments were made with $C_{1-100}H_{4-200}N_{0-4}O_{0-20}S_{0-4}Na_{0-1}Mg_{0-1}Fe_{0-1}B_{0-1}$ as elemental constraints, which yielded thousands of molecular formulas highlighted in Table 1, with less than an 80 ppb root-mean-square error. Although the ultrahigh mass accuracy and isotopic fine structure support confident elemental composition assignments, molecular formulas alone do not distinguish structural isomers or provide compound-specific identifications. Each extraction solvent produced $>18,000$ unique assignments, including heavy isotopologs such as ^{13}C , ^{25}Mg , and ^{26}Mg . The assigned compositions feature up to 20 oxygen, 70 carbon, 4 nitrogen, and/or 4 sulfur atoms. Figure 3 (upper panels) presents the number of assigned molecular formulas for each heteroatom group for the extracted fractions from Murchison. For instance, N_yO_xNa comprises all the formulas with N, O, and Na atoms (e.g., N_1O_5Na or N_2O_2Na). Both

ionization modes reveal significant assignments in the N_yO_x group, as well as many formulas in the O_x and O_xNa heteroatom groups for (−) and (+) ESI. The methanol extract revealed the highest number of formulas in (+) ESI in the N_yO_x group, and in (−) ESI, chloroform assigned the most in this heteroatom class. In addition, there was abundant detection of sodiated N_yO_x adducts in both positive- and negative-ion modes. The O_x class is abundantly present in (−) ESI, whereas the sodiated adducts, O_xNa , are more abundant in (+) ESI. Limited oxygen–sulfur-containing species (S_xO_y) were present in the (+) ESI methanol extract but are abundant in (−) ESI. The number of formulas assigned in the $N_yO_xS_z$ class remains relatively constant across the extractions, except for a spike in the chloroform extract in (+) ESI. In (−) ESI, this trend is not observed, with a decrease in the number of assignments found with each subsequent extract. This suggests that the species observed are more polar in nature as a function of solubility. The number of organomagnesium formulas in the methanol extract shows that these species are preferentially

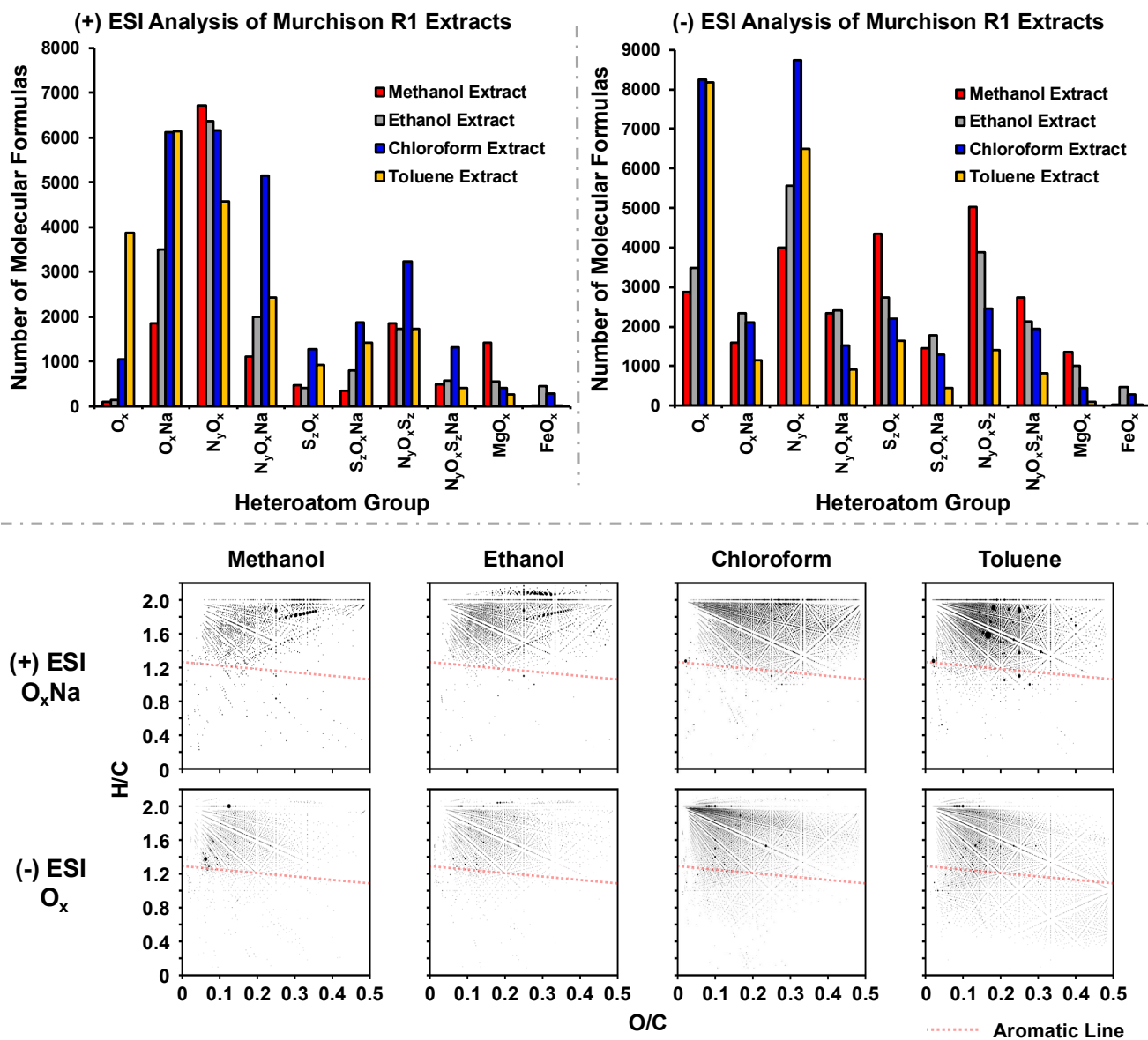


Figure 3. Upper panel: heteroatom-class distribution for the number of molecular formulas assigned in each class for each solvent extract in (+) ESI (left) and (-) ESI (right) from the Murchison meteorite R1 extracts as analyzed by 21T-MS. Lower panel: comparison of O_xNa class from (+) ESI and O_x from (-) ESI for each solvent extract from the Murchison meteorite.

extracted in polar protic solvents, i.e., 2782 assignments combined from (+) and (-) ESI for methanol extract versus 369 assignments for the toluene extract. In addition, hundreds of formulas were assigned to oxygenated iron-containing compounds, a new heteroatom class of organic compounds not yet reported in the SOM of meteorites.

The comparison between HR-AFM image-based structures and ESI FT-ICR MS formula space is used here as a qualitative link between representative imaged molecules and the broader compositional distribution, rather than as a quantitative or exhaustive assessment of PAH abundance. APPI or APCI FT-ICR MS would provide complementary coverage of less polar aromatic hydrocarbons in future studies when sufficient sample is available.

Previous studies into asphaltenic structure have demonstrated the selective extraction of planar PAH structures with acetone as the extraction solvent (M. L. Chacón-Patiño et al. 2020). Utilizing this, an acetone extract of Murchison was

performed after the initial set of methanol through toluene extractions was optimized. Given how little material is yielded during extractions, only (-) ESI FT-ICR MS was collected to allow for enough sample for HR-AFM analysis. From this analysis, 83.0% of the relative abundance was assigned, representing 31,431 of the 35,738 detected peaks. Figure A2(a) shows that the distribution of assignments across the heteroatom groups follows trends seen in the toluene and chloroform extracts. The class with the most assignments is the O_x class, followed by N_yO_x . Like the other extracts, sodiated adducts were present for all the major classes. The major difference in trends observed is the relative lack of organometallic species assigned.

3.3. Visualization of Compositions

van Krevelen diagrams are commonly used for visualizing complex dissolved organic matter analyzed by UHR-MS. These plots are composed of two ratios, the hydrogen-to-

carbon (H/C) ratio on the y-axis and the oxygen-to-carbon (O/C) ratio on the x-axis. Plotting these ratios can make it easier to understand complex mixture composition based on the aromaticity represented by H/C and the oxygen content represented by O/C (M. L. Chacón-Patiño et al. 2015; J. R. Laszakovits & A. A. MacKay 2021; A. M. McKenna et al. 2021).

The trends observed in the van Krevelen distributions are broadly consistent with previous targeted analyses of Murchison and Aguas Zarcas, while extending the accessible molecular space to substantially larger and more complex compounds. Numerous studies have reported amino acids, hydrocarbons, organosulfur compounds, N-heterocycles, and other soluble organic species in both meteorites (L. H. Fuchs et al. 1973; J. R. Cronin & S. Pizzarello 1990; J. C. Aponte et al. 2020; D. P. Glavin et al. 2020; S. Pizzarello et al. 2020; L. D. Tunney et al. 2022; H. Naraoka et al. 2023a). Most of these compounds occur below the lower mass limit of the present measurements (approximately 155 Da) and are therefore largely outside the molecular range investigated here. Nevertheless, the observed enrichment of aromatic formulas in the less polar extracts is consistent with previous reports of PAHs and aromatic hydrocarbons in CM chondrites (J. R. Cronin & S. Pizzarello 1990; K. Slavicinska et al. 2022), whereas the extensive O- and S-bearing molecular populations observed by FT-ICR MS are consistent with the diverse oxygenated and organosulfur chemistries reported for both meteorites (S. Pizzarello et al. 2020; H. Naraoka et al. 2023b).

Rather than targeting individual compound classes, the present 21T-MS measurements provide a comprehensive view of the high-mass molecular inventory, yielding approximately 40,000 assigned molecular formulas across all extracts and ionization modes. This enables detailed comparisons of alkylation, aromaticity, oxygenation, sulfur-containing species, and metal-bearing molecular classes between Murchison and Aguas Zarcas. In particular, the data reveal systematic differences in Fe- and S-containing molecular populations, as well as distinct distributions in H/C and O/C space, providing molecular-level detail that complements and extends previous targeted investigations.

The use of van Krevelen diagrams here allows for the quick determination of the aromaticity of the compounds that were assigned from these extracts. A comparison between the O_xNa class from (+) ESI and O_x in (−) ESI for all four solvent extractions in Figure 3 (lower panel) demonstrates an overlap between the sodium adducts and the acidic species that are commonly ionized in negative-ion mode. This indicates that the sodium adducts do not rely upon random attachment to oxygen-containing compounds but rather interact with acidic groups present. The red dashed line on the plots represents the aromatic line, above which molecules are more aliphatic in nature and below are more aromatic (L. K. McDonough et al. 2022). As can be seen, almost of the formulas assigned sit above the aromatic line, with a majority of the H/C ratios between 1.2 and 2.0 and O/C ratios between 0.1 and 0.5. This indicates that the assigned species are aliphatic in nature. The O_x class van Krevelen plot for the acetone extract (Figure A6) demonstrates a broader range in species, with significantly more aromatic compounds present. The O/C ratios are similar to the other extracts, but the H/C ratios lie between 0.4 and 2.0. As a comparison, natural organic matter found on Earth

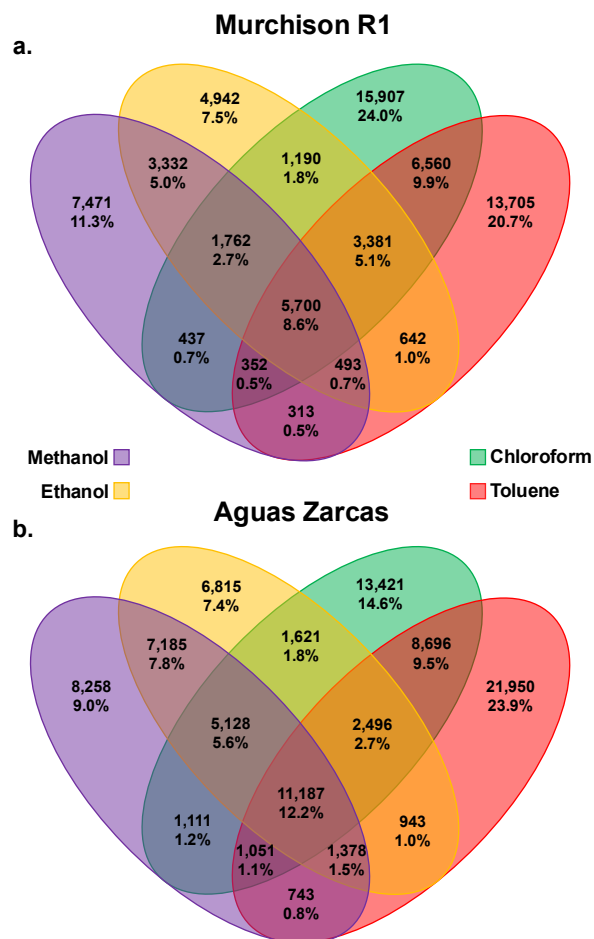


Figure 4. Comparison of assignments made in each solvent extract from Murchison R1 (a) and Aguas Zarcas (b) derived from (+) ESI 21T-MS analysis.

ranges in composition from 0.4 to 1.4 in H/C and 0.3–0.8 in O/C (W. Bahureksa et al. 2022).

Comparison between methanol extracts in Figure A3 for the three Murchison experiments demonstrates both the reproducibility of the extractions and the importance of standardized sample-preparation protocols. The initial method assigned 6330 formulas in the most abundantly assigned group, O_xNa , whereas the first and second replicates for the final method assigned 6713 and 7187 formulas in N_yO_x for (+) ESI. Examination of negative-ion ESI shows that replicates 1 and 2 have 2500 more assigned species in the $N_yO_xS_z$ group compared to the initial method, with the most assigned in O_x at 2735. Both ionization modes yielded at least 600 formulas assigned for organomagnesium species, with 1646 formulas in (+) ESI provided by the initial method and 654 formulas in (−) ESI from R2 of the new method. In addition, only the methanol extraction of the initial method analyzed in (+) ESI displayed any significant assignments in the oxidized iron compounds when compared to the other methods, with over 550 formulas assigned. As seen in Figure 2, however, other solvents extracted oxidized iron species in both ionization modes.

A comparison of formulas assigned with Venn diagrams determines the number of formulas shared between extracts. The revised extraction method of Murchison (Figure 4(a)) shows that only 8.6% of 66,187 unique assigned formulas

Van Krevelen Distributions and ESI Venn Comparisons

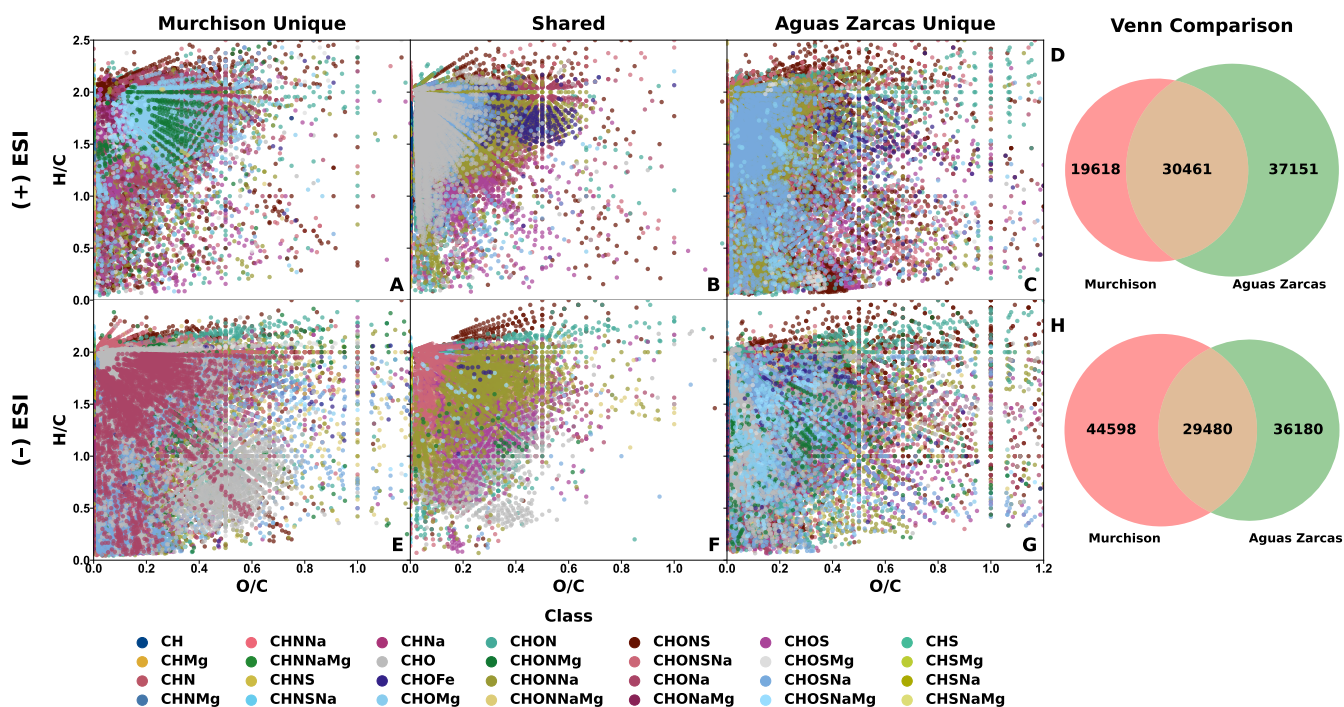


Figure 5. Formula-level comparison of Murchison and Aguas Zarcas SOM based on merged monoisotopic molecular formula assignments from the sequential extracts. Venn diagrams show formulas detected only in Murchison, only in Aguas Zarcas, and shared by both meteorites for (+) ESI and (-) ESI. Corresponding van Krevelen diagrams show the H/C and O/C distributions for each formula group, with heteroatom classes indicated by color. Unique formulas refer to formulas detected only in one meteorite under the present extraction, ionization, and assignment workflow, and should not be interpreted as definitive absence from the other sample.

were shared between the four extracts. Very few formulas were shared between the polar solvents and nonpolar solvents individually. Comparison of the methanol and toluene extracts indicates only 0.5% of the formula assignments are shared (as indicated in the top “Murchi.R1” diagram at the bottom area overlapping area “313, 0.5%”). The polar and nonpolar solvents share more assignments by comparison, with 5.0% between methanol and ethanol and 9.9% between chloroform and toluene. Overall, the new extraction method for Murchison R1 resulted in 66,187 unique assignments for (+) ESI and 92,878 unique assignments for (-) ESI. Figure 4(b) for Aguas Zarcas displays similar trends to those of Murchison. Among all the extractions, only 12.2% of the assigned formulas are shared. Like Murchison, methanol contains more assignments than ethanol, but unlike Murchison, chloroform contains fewer peaks than toluene, which contains nearly 22,000 assigned formulas. Aguas Zarcas overall had 91,981 unique assignments in (+) ESI and 82,783 unique formulas for (-) ESI.

3.4. Comparison of Murchison and Aguas Zarcas

To further compare the soluble organic compositions of Murchison and Aguas Zarcas, all monoisotopic formula assignments from the sequential extracts were merged for each meteorite and compared by ionization mode. Here, “unique” refers to formulas detected only in one meteorite under the present extraction, ionization, and assignment workflow, whereas “shared” refers to exact elemental formulas detected in both meteorites. Therefore, these comparisons should be interpreted as formula-level overlap rather than as absolute presence or absence of specific molecular structures.

In (+) ESI, 30,461 formulas were shared between the two meteorites, whereas 19,618 formulas were detected only in Murchison and 37,151 only in Aguas Zarcas (Figure 5). In (-) ESI, 29,480 formulas were shared, whereas 44,598 formulas were detected only in Murchison and 36,180 only in Aguas Zarcas. Thus, the two CM chondrites contain a substantial common formula-level composition, but each sample also contains a large set of sample-specific formulas. The van Krevelen distributions of the shared and sample-specific formulas show broad overlap across aliphatic, aromatic, and oxygen-rich compositional regions, but the relative density and extent of the distributions differ between the two meteorites and between ionization modes. For comprehensive isolated van Krevelen class distributions, refer to Figures B3 and B4 and Table B1. These differences should not be attributed to solvent extraction alone or to a single parent-body process. Instead, the data are most consistent with the combined effects of extraction selectivity, ionization response, parent-body alteration, mineralogy, sample history, and matrix–organic interactions. This interpretation is particularly important for oxygen- and metal-bearing formulas, including the Mg- and Fe-containing compositions reported here. CM chondrites are mineralogically complex and record aqueous alteration involving hydrated silicates, sulfides, oxides, carbonates, and secondary Fe- and Mg-bearing phases (L. A. Garvie 2021; M. Suttle et al. 2021; I. Kouvatzis et al. 2024). These mineral phases may influence which soluble organic compounds are retained, transformed, or released during extraction. However, the present FT-ICR MS data provide elemental formula assignments only; they do not define the bonding environment of Mg or Fe, nor do they distinguish structural isomers.

Therefore, the comparison between Murchison and Aguas Zarcas shows that their soluble organic compositions are related but not identical at the formula level. The differences are consistent with the known sensitivity of CC organic matter to parent-body alteration, mineral–fluid interaction, and sample history, but they cannot by themselves define a unique alteration pathway. In an astrobiological context, these results do not identify specific biomolecules or demonstrate a direct origin-of-life pathway. Rather, they define a broad chemical space of soluble oxygen-, nitrogen-, sulfur-, magnesium-, and iron-bearing compositions that primitive meteorites could have contributed to early planetary environments.

Mineralogical context is particularly important when interpreting the Fe- and Mg-containing formula assignments. Although both samples are CM chondrites, Murchison and Aguas Zarcas are not mineralogically equivalent. Murchison contains phyllosilicate-rich material with sulfide- and sulfate-bearing alteration products, whereas Aguas Zarcas is a polymict breccia with lithological heterogeneity and reported Fe- and Mg-rich components. These differences may influence the retention, transformation, and extraction of organic compounds associated with mineral surfaces or alteration products. The distinct Fe- and Mg-containing formula distributions observed here are therefore consistent with differences in mineralogical environment, aqueous alteration, and matrix–organic interactions. However, the present data do not establish the coordination environment of Fe or Mg, direct binding to a specific organic structure, or association with an individual mineral phase.

Table B2 lists all identified formula counts for all possible classes for Murchison and Aguas Zarcas.

3.5. Aromaticity Observed

After comparison of solvent-dependent and meteorite-dependent formula overlap, the aromaticity of the assigned species was further evaluated with double-bond-equivalent (DBE; see Equation (1)) versus carbon-number plots.

These plots help establish the aromatic compositional space present in assignments.

$$\text{DBE} = \#C + 1 - \frac{\#H - \#N}{2}. \quad (1)$$

Comparison with DBE versus carbon-number plots of the N_1O_x classes (most abundant class) from each extraction solvent analyzed by (+) ESI for Murchison R2 and Aguas Zarcas is found in Figure 6(a). The shift from methanol to toluene sees an increase in abundance-weighted average DBE for the N_1O_x class from 13.6 to 18.3. The abundance-weighted average carbon number observed for each solvent increases from methanol (35.3) to chloroform (40.3) but then drops in toluene (37.3). This could be the result of the drop in formulas assigned from chloroform to toluene, as seen in Table 1. Aguas Zarcas demonstrates the expected trend of increased DBE from methanol to toluene (9.1–23.5) and an increase in carbon number (36.9–51.2). The ethanol and chloroform extracts, however, did show the same abundance-weighted average carbon number. Figures 6(b) and (c) examine the calculated aromaticity (Equation (2)) and abundance-weighted average O/C ratios of the N_1O_x class for each extract of Murchison R2

and Aguas Zarcas.

$$\text{Aromaticity} = \frac{\text{Abundance-weighted average DBE}}{\text{Abundance-weighted average H/C}}. \quad (2)$$

As seen in Figure 4(a), the aromaticity increases from methanol to toluene for both Murchison R2 and Aguas Zarcas. This indicates that the more nonpolar solvents would extract the more aromatic species, as would be expected. The O/C ratio denotes the amount of oxygen present in the extracted compounds. For Murchison R2, the O/C ratio increases across the extraction solvents, from 0.2 in methanol to 0.4 in toluene. Aguas Zarcas is slightly different in this regard as it peaks in O/C ratio in chloroform before decreasing in toluene. Here, the methanol extract has an O/C ratio of 0.2 for the N_1O_x class, increasing to 0.4 in chloroform before falling to 0.3 in toluene.

These solvent-dependent trends show that meteorite SOM is not adequately represented by a single extract. Targeted analyses have identified numerous classes of soluble organic compounds in carbonaceous meteorites, including amino acids, carboxylic acids, aliphatic hydrocarbons, N-heterocycles, organosulfur compounds, and metal-organic species (J. R. Cronin & S. Pizzarello 1990; A. Ruf et al. 2017; D. P. Glavin et al. 2018; D. N. Simkus et al. 2019; J. C. Aponte et al. 2020; D. P. Glavin et al. 2020; S. Pizzarello et al. 2020; L. D. Tunney et al. 2022). These studies have provided detailed molecular characterization of specific compound families and have revealed differences in the organic inventories of Murchison and Aguas Zarcas. In particular, variations in amino acid abundances, hydrocarbon distributions, sulfur-bearing compounds, and the extent of parent-body processing and aqueous alteration have been reported.

Previous targeted studies provide an important context for the formula-level distributions observed here. Analyses of Murchison and Aguas Zarcas have reported amino acids, amines, carboxylic acids, aliphatic and aromatic hydrocarbons, N-heterocycles, organosulfur compounds, and metal-containing soluble organic species, with sample-dependent differences in their relative abundances and distributions. The enrichment of lower-H/C and higher-DBE formulas in the chloroform and toluene extracts is consistent with recovery of less polar and more aromatic molecular populations, whereas the abundant O-, N-, and S-containing formulas detected by ESI reflect the broad diversity of polar and heteroatom-containing species previously observed in CM meteorites. However, the present FT-ICR MS data provide elemental formulas rather than compound-specific identifications. Individual amino acids, carboxylic acids, PAHs, or sulfur compounds therefore cannot be assigned from these data alone. In addition, because extracts were concentration-normalized before analysis, the number of assigned formulas reflects molecular diversity together with extraction selectivity and ionization response, rather than the concentration of a given molecular family in the meteorite.

In contrast, the present broadband 21T-MS approach targets a substantially different region of molecular space. Owing to the mass range accessible by our measurements, the lower mass limit is approximately 155 Da, corresponding roughly to molecules containing about 12 carbon atoms. Consequently, small and comparatively simple compounds such as amino acids and other low-mass species are largely outside the scope of the present study. Instead, our focus is on larger and more

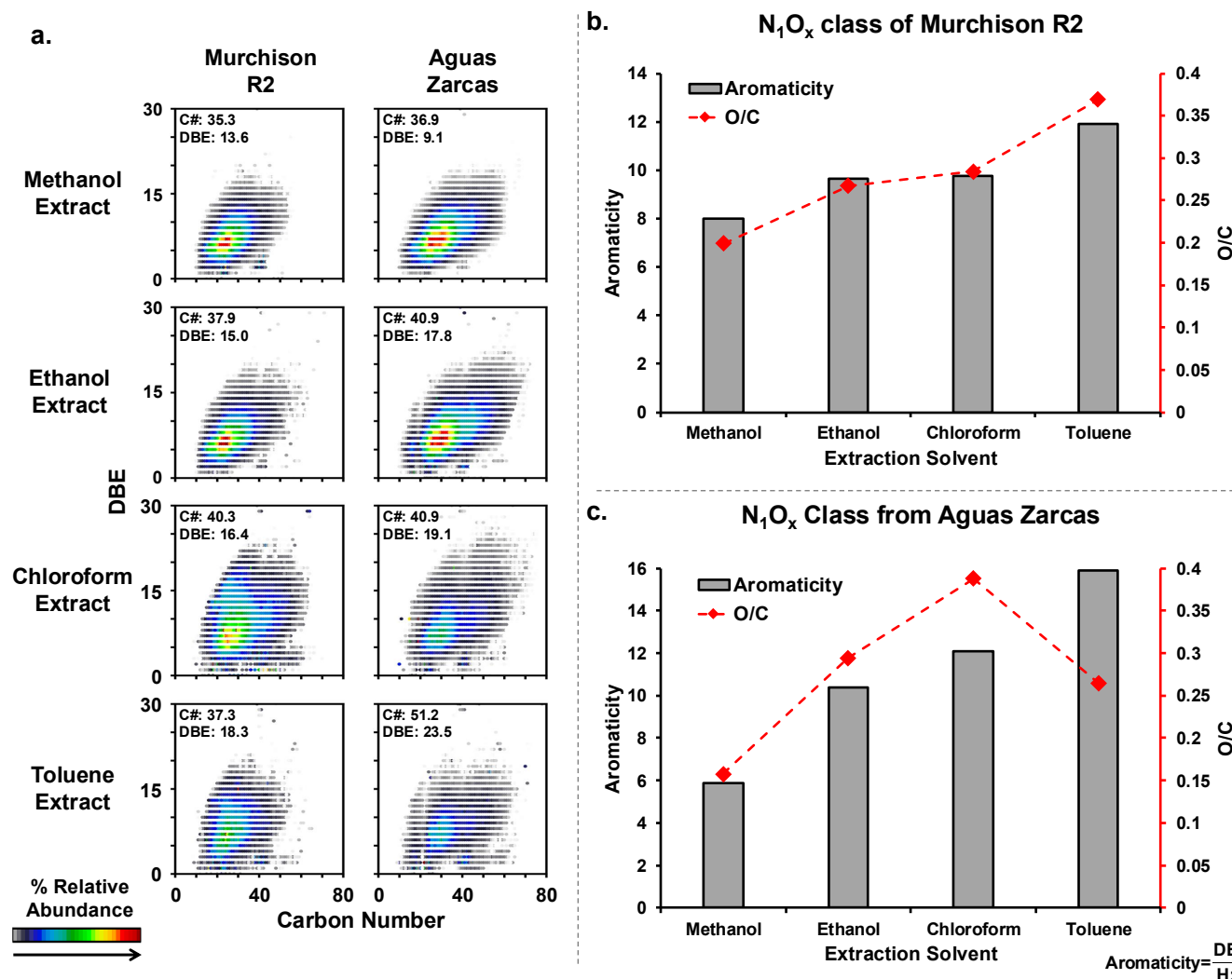


Figure 6. (a) DBE versus carbon-number plots of the N₁O_x class from (+) ESI 21T-MS analysis from Murchison R2 and Aguas Zarcas extractions. (b) Plot of aromaticity and O/C ratios of the N₁O_x class for each extraction solvent from Murchison R2 after analysis by (+) ESI 21T-MS. (c) Plot of aromaticity and O/C ratios of the N₁O_x class for each extraction solvent from Aguas Zarcas after analysis by (+) ESI 21T-MS.

complex organic molecules. While the present approach does not provide compound-specific identifications, it captures the overall molecular formula diversity across a wide mass range, multiple solvent fractions, and complementary ionization modes. In total, approximately 40,000 unique molecular formulas were assigned across all extracts and ionization conditions, including high-mass hydrocarbon species extending to approximately C₇₀, Fe-bearing compounds exceeding C₃₅, and distinct metal-associated molecular populations in Murchison and Aguas Zarcas that have not been reported previously. Furthermore, our dataset enables an unprecedentedly detailed comparison of sulfur-containing and metal-bearing molecular classes across both meteorites, revealing systematic differences in their molecular inventories and metal-organic associations that are not accessible through targeted analyses alone.

The preferential recovery of heteroatom-rich and more aliphatic formulas in polar solvents, and more aromatic formulas in less polar solvents, likely reflects differences in functional-group chemistry, solubility, and interactions with the mineral-organic matrix. The observed differences between Murchison and Aguas Zarcas should therefore be interpreted as the combined result of extraction selectivity, ionization

response, sample history, parent-body alteration, mineralogy, and matrix effects, rather than as evidence for a single controlling process. Together, the targeted studies and the broadband molecular distributions reported here provide complementary views of extraterrestrial organic matter, linking specific compound classes to the broader molecular landscape of carbonaceous meteorites.

Figure A4 displays N_yO_x classes in van Krevelen plots for Murchison replicates and Aguas Zarcas analyzed with (+) ESI. In methanol extracts, most species were assigned to the upper-left corner, which indicates more aliphatic species for both Murchison and Aguas Zarcas. The shift in polarity from methanol to toluene results in different detected species with an increased number of compounds that appear below the aromatic line. This demonstrates that the nonpolar solvents extract the more aromatic species present in each sample. The move from polar to nonpolar is more evident in Aguas Zarcas as the densely populated aliphatic region in methanol shifts to a less populated area in the toluene. Plotting all the oxygen-containing species in Figures A5 and A6, it is possible to compare the differences in van Krevelen plots for positive and negative ESI. With all oxygen-containing formulas included, the x-axis ratios of the van Krevelen plots double from 0–0.5

to 0–1.0 O/C, whereas the y-axis remains relative to what was observed in the N_yO_x van Krevelens plots. Like what was seen in Figure A5, the (+) ESI methanol fractions have a high density in the upper-left corner, above the aromatic line, for both Murchison and Aguas Zarcas. When all the oxygenated formulas are included in a single plot for each extract, more aromatic formulas are present in all plots, including the methanol extracts. The progression from methanol to toluene reveals a persistent trend of increased aromaticity. Negative-ion ESI is slightly different when comparing Murchison and Aguas Zarcas. Across all four solvents, the initial Murchison extractions are distributed similarly, but there are some aromatic shifts in the first and second replicates with changes in solvent composition. Aguas Zarcas displays different trends in the aromaticity present from methanol to toluene. Toluene displays less aromaticity in comparison to the other three solvents, which likely results from the lower number of assignments found in this dataset. The van Krevelen plot of all the oxygen-containing species for the acetone extract demonstrates a slightly different trend in Figure A2(c), with a more densely populated region located between H/C 0.6–1.3. As mentioned previously, (–) ESI UHR-MS has been performed in the past with methanol extractions of Murchison (P. Schmitt-Kopplin et al. 2010; N. Hertkorn et al. 2015; A. Ruf et al. 2017; J. Hertzog et al. 2019). Comparison of the results obtained here to published data indicates similar trends observed between the datasets. However, the methanol extracts from Murchison R1 and R2 shown in Figure A6 do appear to demonstrate a slightly more aromatic nature than the literature data described previously.

3.6. Metal-containing Compositions

Based on the previously reported study by A. Ruf et al. (2017), oxygenated magnesium compounds were detected in a variety of meteorites. Magnesium is a very abundant element in meteorites, typically bound with silicon to form silicon–magnesium–oxygen matrices (K. Lodders 2010). For the observed class of O_4Mg , tandem mass spectrometry experiments provided a base structure of dihydroxymagnesium carboxylates, where a $Mg(OH)_2$ group detaches from the matrix when heated and coordinates with existing carboxylic acids (A. Ruf et al. 2017). Given the previous work that established magnesium as a key element of interest in Murchison and the importance of magnesium in living systems, this element was taken into account for the data analysis for both meteorites.

The Mg- and Fe-containing formula assignments are consistent with the mineral-rich and aqueously altered nature of CM chondrites, but their exact coordination environments cannot be defined from formula-level FT-ICR MS alone (L. A. Garvie 2021; M. Suttle et al. 2021). For detailed metal-bearing formula comparisons, refer to Section B.1 and Figure B1.

Murchison R1 had the most organomagnesium formulas assigned out of the Murchison extractions, with thousands of formulas for species across all four extraction solvents in both (+) ESI (2641) and (–) ESI (2929). Aguas Zarcas, however, did not contain many oxygenated magnesium assignments, with only 443 assignments in (+) ESI and 679 in (–) ESI across all four solvents. In Murchison, methanol extracted the most species, whereas in Aguas Zarcas, chloroform did. This may indicate that the little organomagnesium that was present

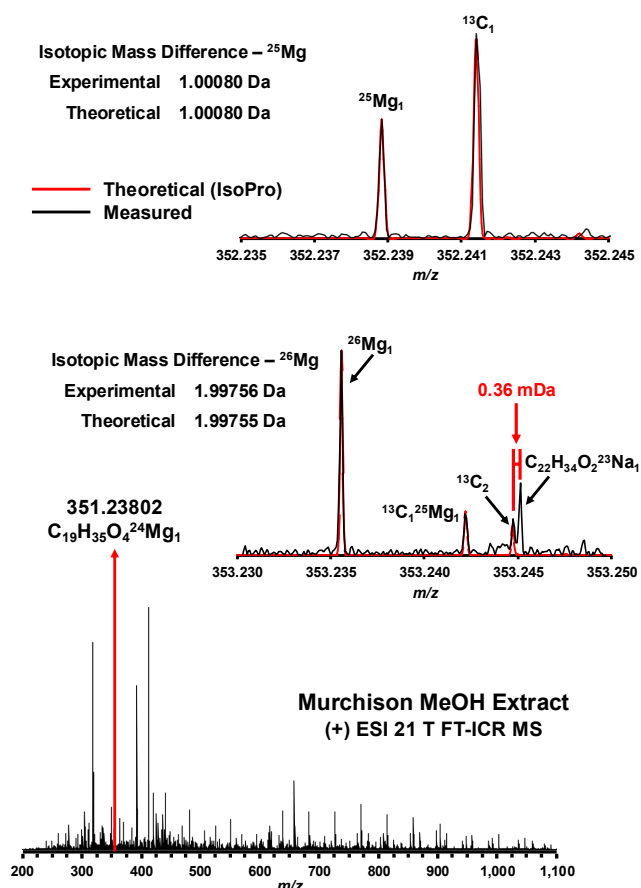


Figure 7. From a methanol extract of Murchison analyzed by (+) ESI FT-ICR MS, the peak at m/z 351.23802 was assigned as $C_{19}H_{35}O_4^{24}Mg_1$, displaying the isotopic fine-structure peaks of ^{25}Mg and ^{13}C (top) and ^{26}Mg , $^{13}C^{25}Mg$, and $^{13}C_2$ (bottom) in both the theoretical and measured fine structure. Also detected is $C_{22}H_{34}O_2^{23}Na_1$, which is separated from the $^{13}C_2$ peak by 0.36 mDa.

in Aguas Zarcas is more aromatic in nature compared to the more aliphatic species in Murchison. The comparison of van Krevelen plots (see also Section 3.3) for each meteorite's O_xMg classes in Figure A7 reflects what was seen in the assignments. Murchison has significantly more data points, specifically assignments between $O_{4-9}Mg$ present in the plot, with most of the points above the aromatic line, indicating an aliphatic nature. Aguas Zarcas contains very few points, but the distribution is much more even between the aliphatic and aromatic regions.

An extensive analysis and comparison of sulfur- and metal-bearing molecular populations, including CHOF_e and CHOMg species, between Aguas Zarcas and Murchison is presented in Appendix B.

To confirm these assignments, the isotopic fine structure of oxygenated magnesium compounds can be compared against theoretical values calculated with IsoPro 3.1. One example from (+) ESI analysis of Murchison MeOH extract is demonstrated in Figure 7, examining $C_{19}H_{35}O_4^{24}Mg_1$. There are three major isotopes of magnesium: ^{24}Mg , ^{25}Mg , and ^{26}Mg . The main isotope, ^{24}Mg , has an abundance of 78.99%; ^{25}Mg has an abundance of 10.00%; and ^{26}Mg has an abundance of 11.01%. Both other isotopes of magnesium can be easily identified, and their isotopic fine structure matches the theoretical fine structure, as demonstrated in Figure 7. In

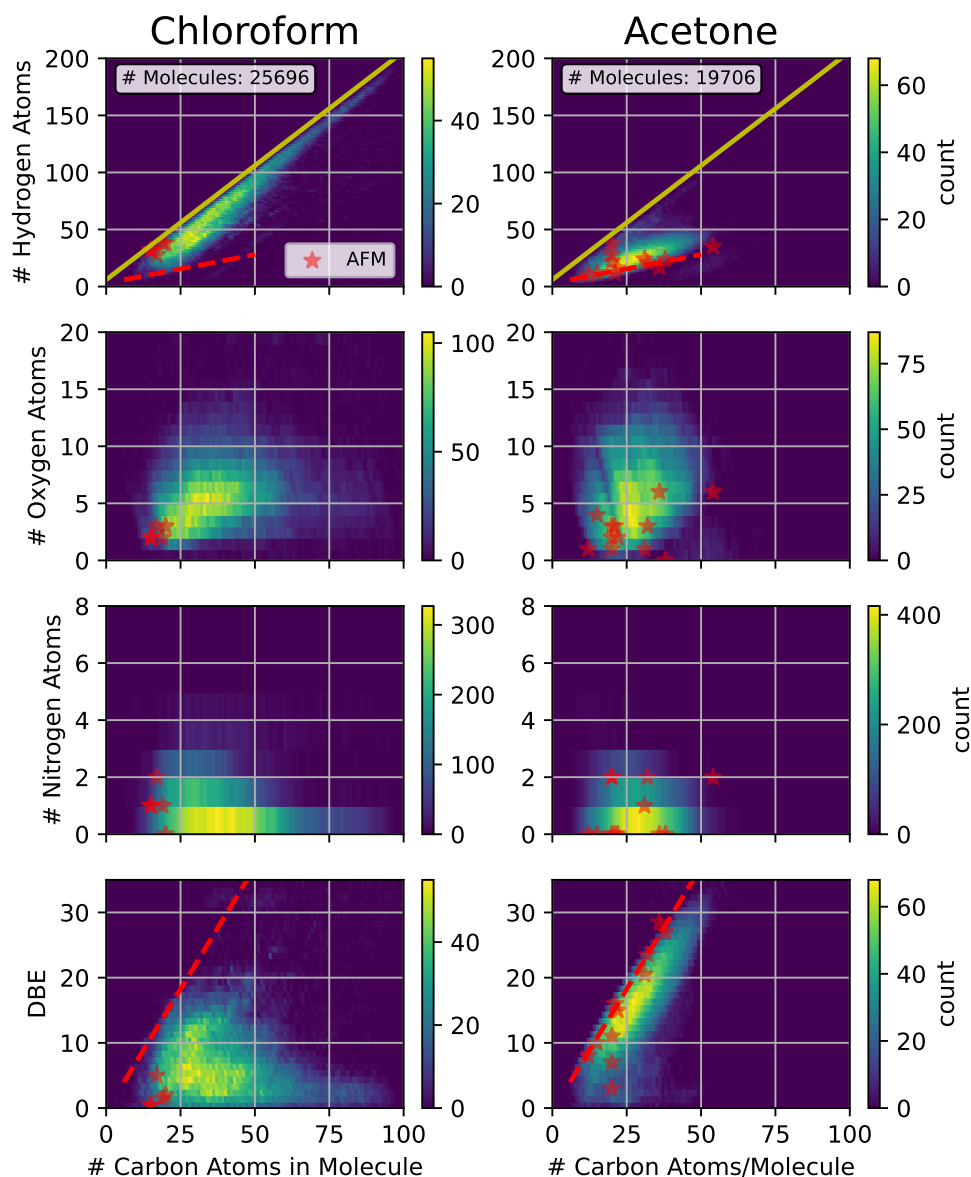


Figure 8. Overlay of estimated structural motifs identified by AFM and the corresponding locations in relation to the HR-MS data for the chloroform and acetone extracts. The plots presented here are for hydrogen, oxygen, and nitrogen number versus carbon number, in addition to the double bond equivalent (DBE) versus carbon number, from the analysis of (–) 21T-MS data. The stars on each plot highlight the potential molecular formula locations for the estimated structures. The data behind this figure are available via doi:[10.5281/zenodo.18891055](https://doi.org/10.5281/zenodo.18891055).

addition, ^{13}C isotopes confirm these assignments, with ^{13}C , $^{13}\text{C}^{25}\text{Mg}$, and $^{13}\text{C}_2$ isotopes assigned and in agreement with the theoretical fine structure. The ultrahigh resolving power of 21T-MS enables near-baseline resolution of the $^{13}\text{C}_2$ peak from an entirely different species, $\text{C}_{22}\text{H}_{34}\text{O}_2^{23}\text{Na}_1$, with an incredibly tight mass split of 0.36 mDa. This underscores the necessity of the 21 T system for attaining such a very high performance. FT-ICR MS performed at lower magnetic fields would not be able to resolve a mass split that is subelectron in mass.

3.7. HR-AFM Imaging

The complementary nature of HR-AFM and FT-ICR MS has resulted in multiple studies that explore the structural motifs in complex mixtures, such as petroleum by B. Schuler et al. (2017a) and meteorite extracts by K. Kaiser et al. (2022). These pair UHR-MS data with snapshots of molecular

structures obtained by HR-AFM to provide more insight into the structural motifs that are observed in the UHR-MS data. The previous meteorite MS-AFM study focused on imaging a chlorobenzene extract, along with the unextracted and residue material, to compare with LDI data collected on a 12T FT-ICR MS system.

For this study, material from the chloroform and acetone extracts of Murchison was sent to Brookhaven National Laboratory for HR-AFM imaging. The molecular formulas of potential molecular motifs observed are overlaid on HR-MS data obtained from negative ESI in Figure 8. Here, plots of the number of hydrogen, oxygen, nitrogen, and the DBE versus carbon number are presented. As discussed previously, the chloroform extract demonstrates more aliphatic-type material, while the acetone is more aromatic. When looking at the distribution of AFM structures for chloroform, this is supported. This is further explored in Figures 9 and 10, where images of the observed structures are presented. The top and

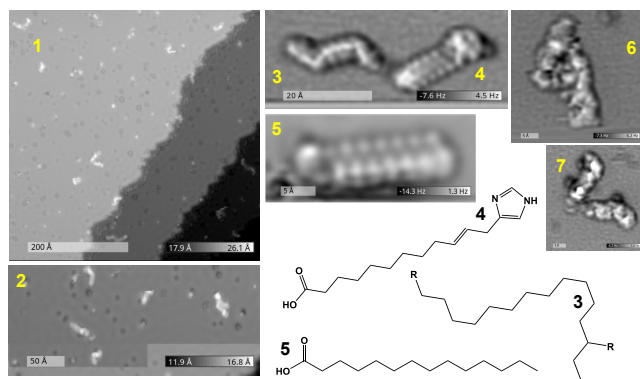


Figure 9. Presented here is HR-AFM molecular imaging performed on the chloroform extract of the Murchison meteorite. Image 1 gives a broad view of the Cu(111) crystal with molecular motifs present in STM mode. The zoom in of image 2 provides more detail than the larger overview. HR-AFM images 3–5 show aliphatic, saturated hydrocarbon chains with R groups present at either end. Approximate structures are shown below these images. Images 6 and 7 highlight the difficulties in performing these measurements, where structural resolution was unobtained, even at the same distances and approximate frequencies as those for other resolved structures.

bottom plots show the aromaticity of the structures observed. The hydrogen versus carbon plot shows the broader distribution of structures in the acetone extract, while the chloroform is more concentrated in the lower hydrogen/carbon-number region. A similar trend is seen in the DBE versus carbon-number plot, with the acetone structures falling along the planar PAH line, indicative of highly aromatic planar structures. The second and third rows of plots show the distribution of heteroatoms found in the AFM structures. The AFM structures observed contain a lower heteroatom distribution compared to the HR-MS data, with the chloroform structures concentrated in the low-carbon-number, low-oxygen/nitrogen regions. However, the acetone extract exhibits a broader distribution of carbon numbers and oxygenated structures. The reduced presence of heteroatom-rich species is expected, as such compounds are challenging to image and rarely yield unambiguous structural information.

Figure 9 presents multiple images from the chloroform extract analysis. Images 1 and 2 give a broad view of the material deposited on the Cu(111) crystal. Observed in these two images are multiple structures with chainlike features. The zoom ins of images 3–5 show these aliphatic structures with R groups of various kinds. Below these are approximate structures for each of the highlighted images. Image 3 shows a long, aliphatic, saturated chain (B. Schuler et al. 2017b) with a potential R group at one end and another branched R group toward the opposite end. These potential R groups include $-H$, $-NH_2$, and $-OH$. Image 4 provides more variation in structure and R groups. Here, a straight chain with a potential carboxylic acid group at one end and an imidazole group at the other is shown, with a double bond two carbons away. The double bond is estimated due to the drop in frequency in that region. Image 5 shows the clearest image of a saturated, straight chain with an R group, most likely a carboxylic acid, at one end. The other end terminates in a methyl group. The remaining images 6 and 7 highlight the difficulty in performing these measurements. These are two structures that cannot be resolved properly. Image 6 may have contained a ring system, while 7 could have contained a carbon chain. However, without a clearer image, it is impossible to say with any form of

certainty. This mainly points to highly nonplanar molecules with complex 3D shapes, likely also including many heteroatoms, versus an aromatic carbon core.

As seen in Figure 8, the acetone extract provided significantly more aromatic structures for imaging. Found in Figure 10 are 14 examples of these structures, with potential structures for each shown below. These molecular motifs span a range of species from long, saturated chains to condensed aromatic cores. Image 1 demonstrates an unusual structure very similar to that of archipelago asphaltene, with small aromatic cores connected by alkyl linkages. There are multiple R groups present, with one estimated to be a carboxylic acid group. The others could be $-CH_3$, $-OH$, or $-NH_2$ groups, as these are smaller functional groups that are more difficult to resolve. The second image presents a smaller molecule with a continuous core structure. One side shows a higher-frequency response, which was tentatively drawn as carboxylic acid, hydroxyl, and methyl groups. The other end shows a potential pyridazine ring system due to the drop in frequencies. Image 3 begins to show a longer ring system with three potential carboxylic acids groups. Image 4 has similar characteristics compared to image 2, with a central core structure, a carboxylic acid group off one end, and two R groups, which may be $-CH_3$ and $-NH_2$ based off their size. Images 5 and 6 demonstrate similar structures with long ring chains of aromatic species. Both contain potential nitrogen-containing rings, pyrrole and pyrrolidine, as part of the backbone of rings. Image 5 has an R group at one end that is well resolved but is of unknown type so far, while image 6 has a very well-resolved benzene attached. The smaller ring system found in image 7 contains four aromatic rings connected to an aliphatic ring with multiple groups attached. These include two unresolved R groups and a potential ester. Image 8 contains another long aromatic chain with multiple R groups and a naphthalene group containing an additional R group. The upper R group could potentially be an ethyl group, but the other two R groups are unresolved. The next two images 9 and 10 both have four-ring aromatic core systems. Image 9 has a saturated cyclopentane with an unresolved R group attached. Image 10 contains a benzene attached with a hydroxyl group and a potential amine off the main core structure. Image 11 looks more like those observed in the chloroform extract, with a long saturated chain with two R groups attached. The lower one may be a ring system, while the other is not completely resolved. The remaining three images demonstrate much larger systems. Image 12 contains similar characteristics to image 1, with an archipelago-style central structure. Present off the core structure are carboxylic acid and hydroxyl groups, in addition to two unresolved R groups. Image 13 shows a more centralized, island-type structure with saturated rings fused with aromatic rings. A potential aldehyde group is observed here, the first seen in these structures, along with three unresolved side groups. The largest central core structure is found in image 14, with a similar fused system between saturated and aromatic six-membered rings. There is a single unresolved R group present, which may be a hydroxyl or methyl group. The bottom of image 14 also shows some of the additional difficulties in imaging these types of molecules. The long, black, striated bar is where a molecule became attached to the tip of the probe, dragging across the image. This cut off the image of the molecule present in image 14, therefore leading to an incomplete structure. In addition to these 14

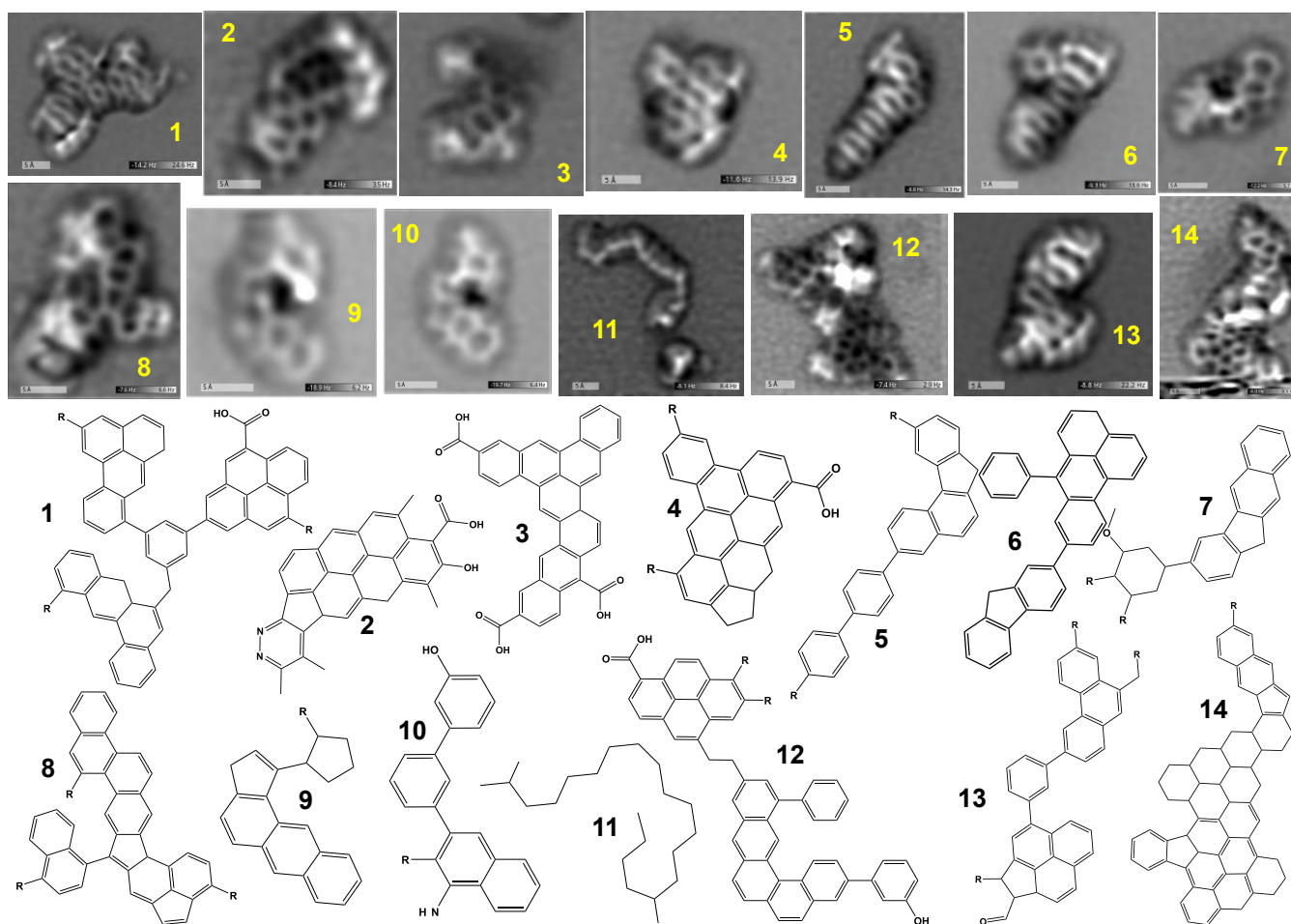


Figure 10. Presented here are HR-AFM images of molecules found in the acetone extract of Murchison with potential structures provided beneath. The light gray scale bar shown on the left in each image is 5 Å long. The image grayscale represents the frequency shift (typical range: -10 Hz, ..., $+5$ Hz), where brighter (more positive shift) means a more repulsive force is detected, whereas darker (more negative shift) means an attractive force is acting on the CO probe. The $-R$ side groups of each range from $-H$, $-CH_3$, $-OH$, $-NH_2$, etc. While key core structures are clear, uncertainty remains about nonplanar side groups at the edges, which appear mostly very bright (partially out of the molecular core plane).

images, there are four further images and structures found in Figure C2, which have similar structural features to those observed in Figure 10. It is important to note that the potential structures found in Figures 9, 10, and C2 are just that: potential. For definitive structures to be identified, model compounds of each would also have to be imaged to provide a certain match. We do have for a subset of molecules shown in Figure 10 (images 1, 2, 4, 5, 6, and 13) limited force-volume maps (supplemental Figures C3, C4, and C5). While early “machine learning” models are being worked on to computationally deduce chemical structures from such datasets, to date reliable analysis methods are not available (J. Carracedo-Cosme et al. 2023; J. Carracedo-Cosme & R. Pérez 2024).

4. Conclusions

Ultra-high-resolution 21T-MS combined with sequential solvent extraction enabled the assignment of unprecedented numbers of molecular formulas across a wide range of organic classes in both Murchison and Aguas Zarcas. Because this study employed nontargeted broadband FT-ICR MS, the results are not intended to provide compound-specific identification of small prebiotic molecules such as amino acids or nucleobases, which require dedicated extraction,

chromatographic separation, authentic standards, and MS/MS-based confirmation. Instead, the present work focuses on the larger and more complex molecular fraction of SOM. The combination of polarity-graded extraction, (+) and (−) ESI, and ionization modifiers resulted in the most comprehensive molecular coverage reported to date for these meteorites. The observed solvent-dependent shifts in aromaticity demonstrate that no single extract adequately represents meteorite SOM, with polar solvents preferentially recovering more aliphatic and heteroatom-rich species and nonpolar solvents enriching larger aromatic compounds.

Across all extracts and ionization modes, approximately 66,000 (+ESI) and 93,000 (−ESI) unique molecular formulas were assigned for Murchison, compared to approximately 92,000 (+ESI) and 83,000 (−ESI) for Aguas Zarcas. Despite both meteorites belonging to the CM2 class, only a minority of formulas are shared between them, indicating substantially different molecular inventories. The corresponding van Krevelen diagrams reveal distinct H/C and O/C distributions, including systematic differences in aromatic, oxygenated, sulfur-bearing, and metal-associated molecular populations. These compositional differences, together with the distinct CHOFe and CHOMg distributions observed for both

meteorites, provide insight into the different parent-body alteration histories, mineralogical environments, and metal-organic interactions that shaped their organic inventories.

The observation of organomagnesium compounds showed trends similar to those of other oxygenated metal-bearing molecular classes, suggesting that related metal-organic complexes may exhibit comparable chemical behavior. In addition to CHOMg species, the present work reveals extensive populations of Fe-bearing compounds, including molecular formulas extending to approximately C_{35} . The distinct CHOFe and CHOMg distributions observed in van Krevelen space and carbon-number analyses suggest chemically meaningful molecular populations rather than isolated assignments. Furthermore, systematic differences between the Fe- and Mg-associated molecular populations of Murchison and Aguas Zarcas are consistent with previously reported differences in mineralogy, aqueous alteration history, and the occurrence of Fe- and Mg-rich lithologies in these meteorites. Future studies of IOM using 21T-MS may reveal additional oxygenated metal-organic complexes and further clarify the relationship between mineral phases and extraterrestrial organic chemistry.

HR-AFM provided single-molecule images that resolve actual chemical structures, thereby complementing the extensive molecular formula assignments from mass spectrometry with direct structural ground-truth examples. The more nonpolar a solvent, the more aromatic the extracted species, ranging from saturated aliphatic chains to highly condensed aromatic cores. Together, the unprecedented molecular coverage achieved by 21T-MS and the direct structural information obtained by HR-AFM demonstrate that primitive solar system materials preserve an exceptionally rich inventory of high-mass organic matter, including hydrocarbons and oxygenated species extending toward C_{70} . The striking molecular differences observed between Murchison and Aguas Zarcas indicate that organic evolution is strongly influenced by parent-body alteration, mineralogical environment, and metal-organic interactions.

The molecular differences observed between Murchison and Aguas Zarcas are best interpreted as evidence that SOM in CM chondrites reflects both inherited chemistry and subsequent parent-body processing. The solvent-dependent shifts in aromaticity and heteroatom content, together with the distinct distributions of sulfur-, Fe-, and Mg-containing formulas, are consistent with variable mineralogical environments, aqueous alteration histories, and matrix-organic interactions. These data do not isolate a single controlling process, identify individual prebiotic molecules, or establish a direct origin-of-life pathway. Instead, they define a broad molecular inventory through which mineral evolution and organic chemistry may

have remained coupled during the early history of primitive solar system materials.

Acknowledgments

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This manuscript has been peer-reviewed by two independent experts.

Software: PetroOrg (Y. E. Corilo 2014), Gxsm (P. Zahl & T. Wagner 2026).

Author Contributions

J.F. and M.C. performed meteorite material curation, extractions, and MS measurements and analysis. M.C. and J. F. were responsible for writing the manuscript and preparing the figures. P.Z. carried out the final sample preparation for HR-AFM, performed the HR-AFM experiments and image analysis, conducted chemical structure assignments together with J.F. and M.C., structure-to-mass estimations and visualizations, contributed to structural interpretation and discussion, writing of AFM-related sections of the manuscript, and finalizing, revising, formatting, and submission of the manuscript. A.M. and R.R. supported the FT-ICR MS operation.

Data Availability

Data for selected figures (via MRT files and tables) and additional files are available via doi:[10.5281/zenodo.18891055](https://doi.org/10.5281/zenodo.18891055).

The authors declare no competing financial interests.

Appendix A

Additional FT-ICR MS Data and Figures

A.1. Comparison of Ion Depletion Off versus On

Figure A1 shows complementary mass spectra of the Murchison ethanol extract.

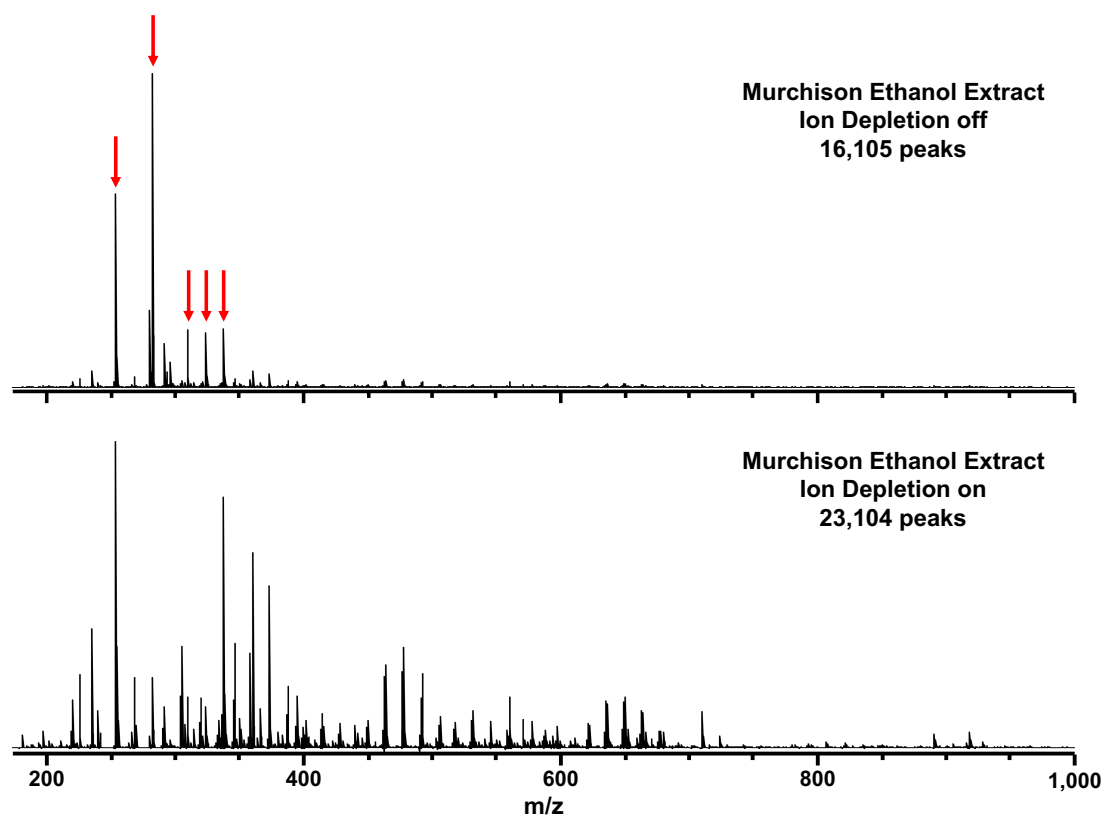


Figure A1. Murchison ethanol extraction without (top) and with (bottom) ion-depletion waveform applied to abundant chemical contaminants as collected in (+) ESI 21T-MS.

A.2. Analysis of the Acetone Extract from Murchison

Figure A2 shows complementary acetone extract heteroatom-class assignments and van Krevelen diagrams.

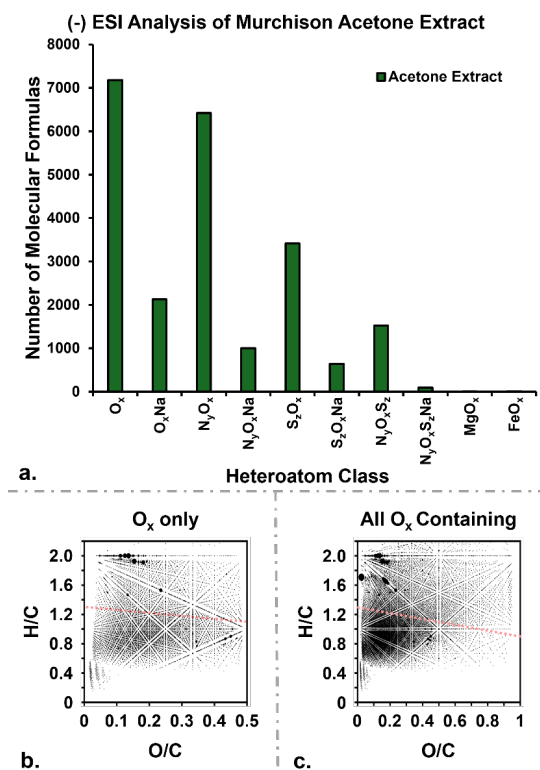


Figure A2. (a) Heteroatom-class distribution for the number of molecular formula assignments in each class as revealed by (–) ESI 21T-MS analysis from the Murchison acetone extract. (b) van Krevelen diagram of the O_x class for the Murchison acetone extract. (c) van Krevelen diagram of all oxygen-containing classes from the Murchison acetone extract.

A.3. Comparison of Heteroatom Distribution across Methanol Extracts for Murchison

Figure A3 shows complementary heteroatom distributions across methanol extracts for Murchison.

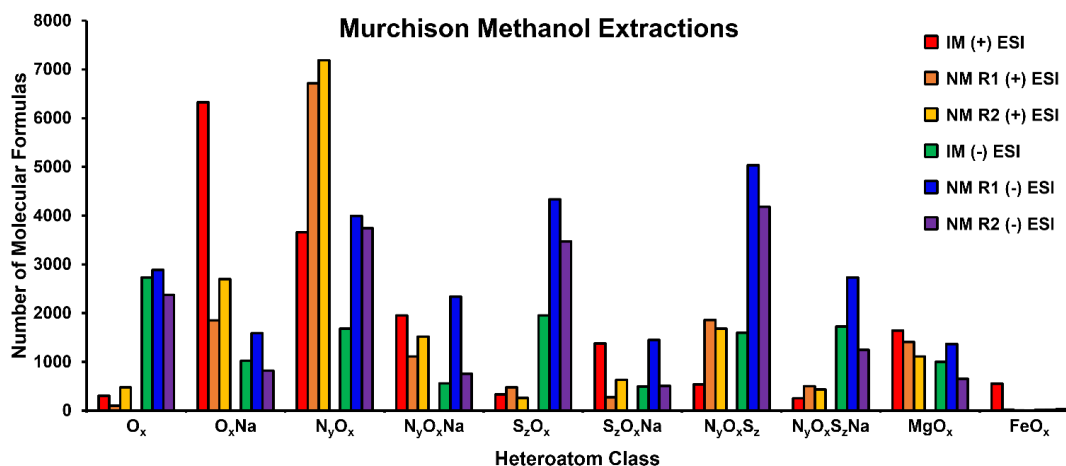


Figure A3. Heteroatom-class distribution of methanol extracts from the initial extraction method and the final-method replicates 1 and 2 from Murchison, analyzed by (+/-) ESI 21T-MS.

A.4. van Krevelen Diagrams of Assigned NyOx Classes in (+) ESI

Figure A4 shows complementary van Krevelen diagrams comparing the assigned NyOx classes from the three Murchison extractions.

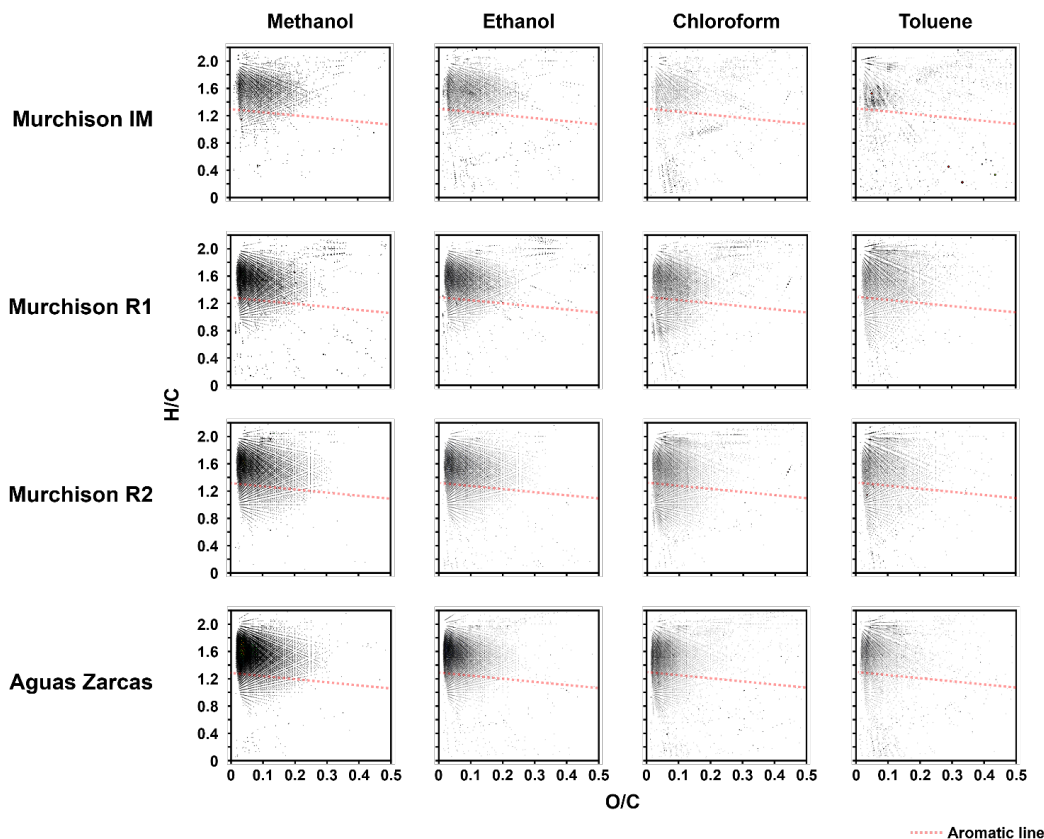


Figure A4. van Krevelen diagrams comparing the assigned NyOx classes from the three Murchison extraction series and Agua Zarcas in each extraction solvent derived from (+) ESI 21-MS.

A.5. *van Krevelen Diagrams of All Assigned Ox Classes in (+/-) ESI*

Figures A5 and A6 show complementary van Krevelen plots of each extraction solvent from Murchison and Aguas Zarcas for (+/-) ESI.

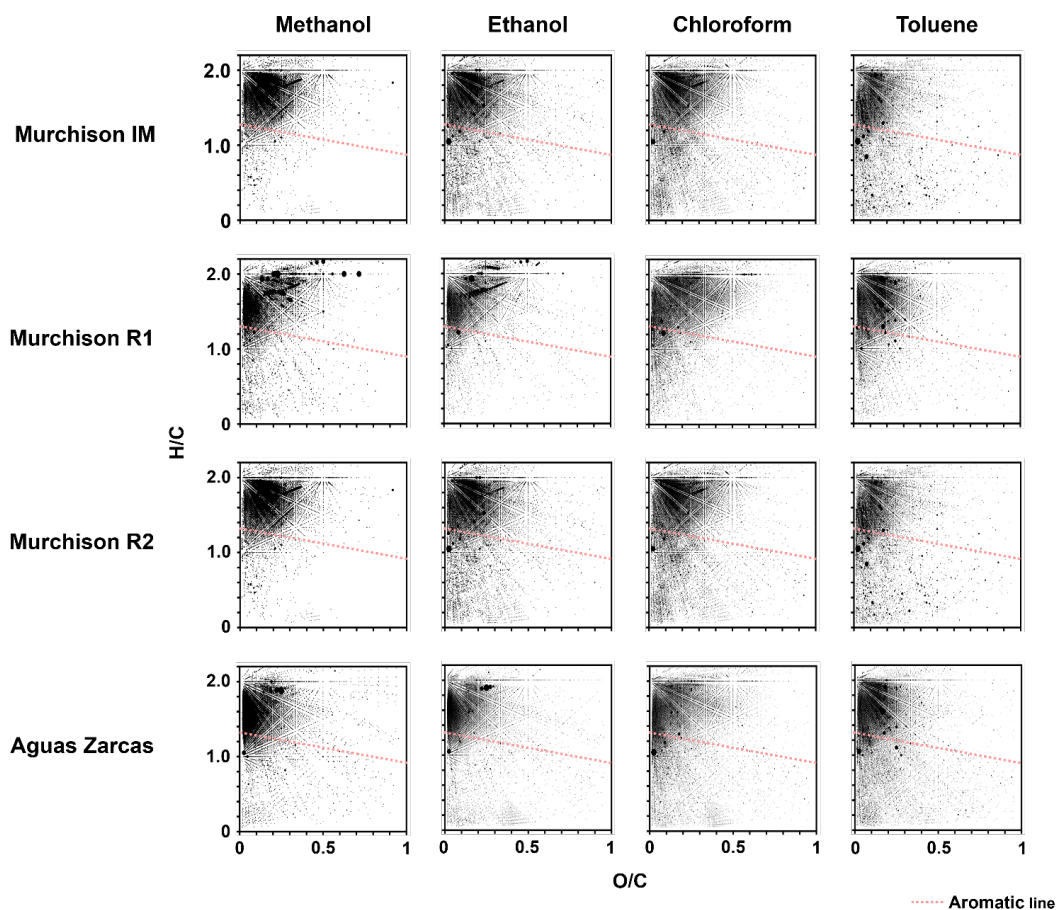


Figure A5. van Krevelen plots of each extraction solvent from the three Murchison extraction series and Aguas Zarcas derived from (+) ESI 21T-MS analysis.

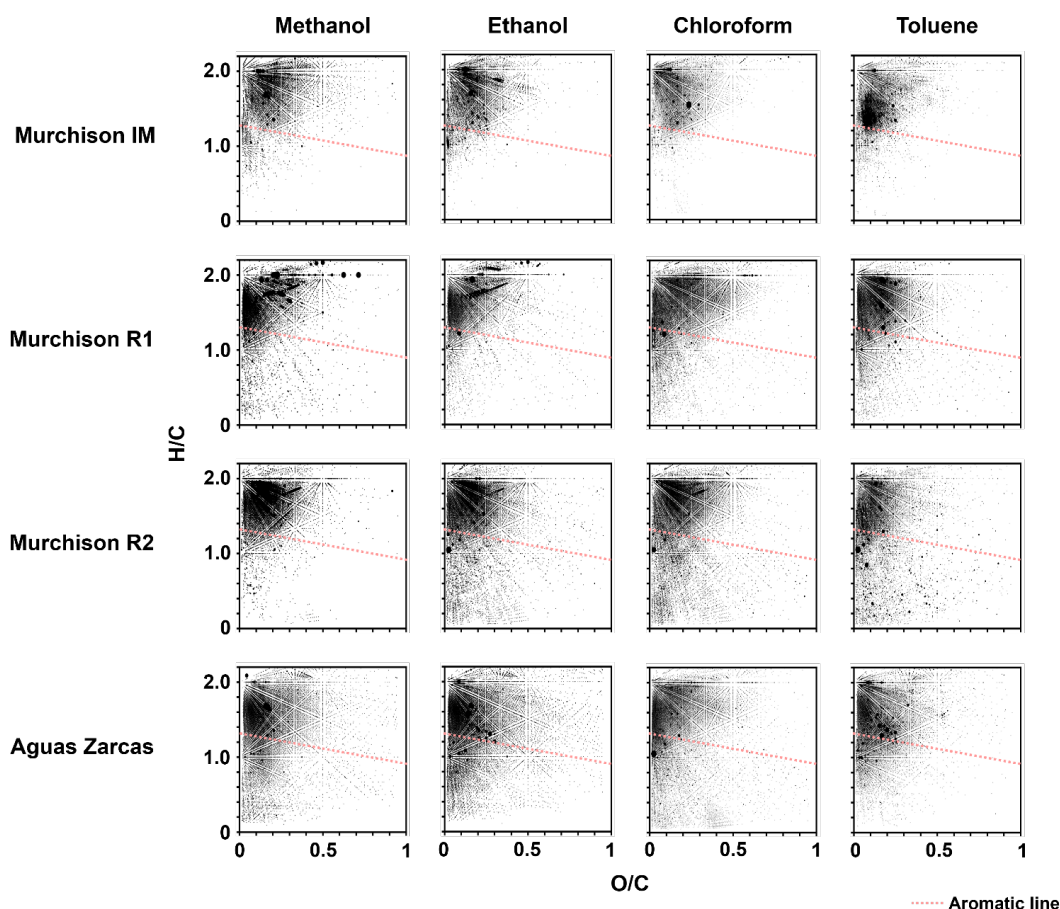


Figure A6. van Krevelen plots of each extraction solvent from the three Murchison extraction series and Aguas Zarcas derived from (–) ESI 21T-MS analysis.

A.6. van Krevelen Diagrams of OxMg Assigned in (+/–) ESI

Figure A7 shows complementary van Krevelen diagrams of the OxMg classes.

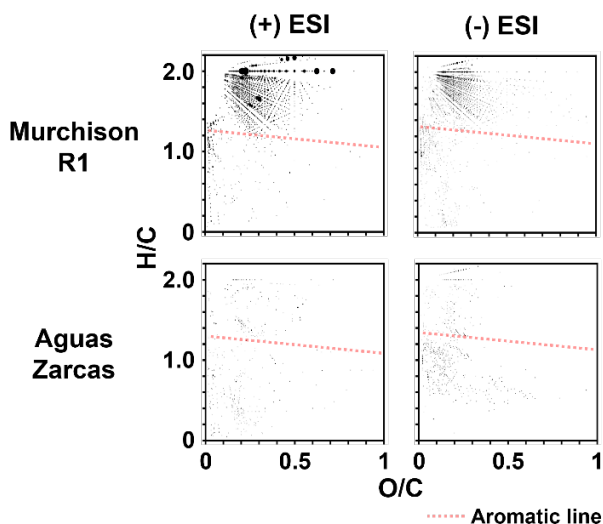


Figure A7. Comparison of OxMg class from (+) ESI and (–) ESI from the Murchison and Aguas Zarcas meteorites.

Appendix B

Exhaustive Statistics of Combined MS Data for Murchison and Aguas Zarcas

B.1. Comparisons of All Combined Metal-bearing (in Particular Fe) Formulas Found in Murchison versus Aguas Zarcas

To study and compare the metal-bearing hydrocarbon compounds found in Murchison and Aguas Zarcas, we isolated molecular formulas containing Fe, Mg, Na, as well as S, and constructed van Krevelen diagrams, as shown in Figure B1. To further assess the compositional range in terms of molecular size, we analyzed the distributions of H, O, and N atom counts, as well as the resulting DBE, as a function of carbon number panel (C) with Fe in Figure B2.

Of particular relevance are the Fe classes: The Fe-containing (Figures B2 panel (C) with Fe and B1 for related van Krevelen plots) molecular formulas with high carbon contents up to C_{35} exhibit a clear bimodal distribution in Murchison, with distinct clusters at ($H/C \approx 2$, $O/C \approx 0.27$) and ($H/C \approx 0.8$, $O/C \approx 0.5$), which are notably offset from the main non-Fe organic population centered around $H/C \approx 1.8$ and $O/C \leq 0.25$, as commonly observed for CM chondrite SOM (P. Schmitt-Kopplin et al. 2010). Although a weaker signal remains near this baseline region, Fe-associated species are

Table B1
Murchison Metal Compounds with Mg

C	H	N	O	S	Na	Mg	Fe	Formula	HC	OC
5	9	0	4	1	0	1	0	C5H9O4S1Mg1	1.800000	0.800000
5	11	0	5	0	0	1	0	C5H11O5Mg1	2.200000	1.000000
5	11	0	5	0	1	1	0	C5H11O5Na1Mg1	2.200000	1.000000
6	11	0	3	1	0	1	0	C6H11O3S1Mg1	1.833333	0.500000
6	13	0	5	0	0	1	0	C6H13O5Mg1	2.166667	0.833333
6	14	1	6	0	0	1	0	C6H14N1O6Mg1	2.333333	1.000000
6	15	0	4	0	0	1	0	C6H15O4Mg1	2.500000	0.666667
6	15	0	5	0	0	1	0	C6H15O5Mg1	2.500000	0.833333
6	15	0	7	1	0	1	0	C6H15O7S1Mg1	2.500000	1.166667
7	13	0	17	0	0	1	0	C7H13O17Mg1	1.857143	2.428571

Note. The first 10 lines of MRT-Table file Murchison-Mg.mrt.txt in doi:[10.5281/zenodo.18891055](https://doi.org/10.5281/zenodo.18891055). The tables containing the Murchison metal compounds and Aguas Zarcas compounds with Mg, Na, Fe, S, C, N, and O are also available via doi:[10.5281/zenodo.18891055](https://doi.org/10.5281/zenodo.18891055).

Table B2
Number of Formulas of All Classes Identified as of Combined (+/-) ESI-MS Data

Class	# in Murchison	# in Aguas Zarcas
CHNO	9796	13826
CHONa	8493	9637
CHNONa	6698	9904
CHNOS	6052	9931
CHO	3817	2962
CHOSNa	3064	4819
CHOS	2695	4100
CHNOSNa	2351	5287
CHOMg	1667	491
CHN	1487	2550
CHOFe	764	1065
CHOSMg	756	83
CHNS	673	1508
CH	416	419
CHNOMg	382	0
CHNSNa	365	613
CHSNa	98	193
CHS	90	55
CHOMgNa	89	0
CHNNa	88	102
CHOSMgNa	72	0
CHMg	55	34
CHNa	36	28
CHSMgNa	21	0
CHNMg	19	0
CHSMg	17	5
CHNOMgNa	13	0
CHNMgNa	5	0

The following classes have not been detected at all:

CHFe, CHNaFe, CHMgFe, CHMgNa, CHMgNaFe,
 CHSFe, CHSNaFe, CHSMgFe, CHSMgNaFe,
 CHONaFe, CHOMgFe, CHOMgNaFe,
 CHOSFe, CHOSNaFe, CHOSMgFe, CHOSMgNaFe,
 CHNFe, CHNNaFe, CHNMgFe, CHNMgNaFe,
 CHNSFe, CHNSNaFe, CHNSMg, CHNSMgFe,
 CHNSMgNa, CHNSMgNaFe,
 CHNOFe, CHNONaFe, CHNOMgFe, CHNOMgNaFe,
 CHNOSFe, CHNOSNaFe, CHNOSMg, CHNOSMgFe,
 CHNOSMgNa, CHNOSMgNaFe.

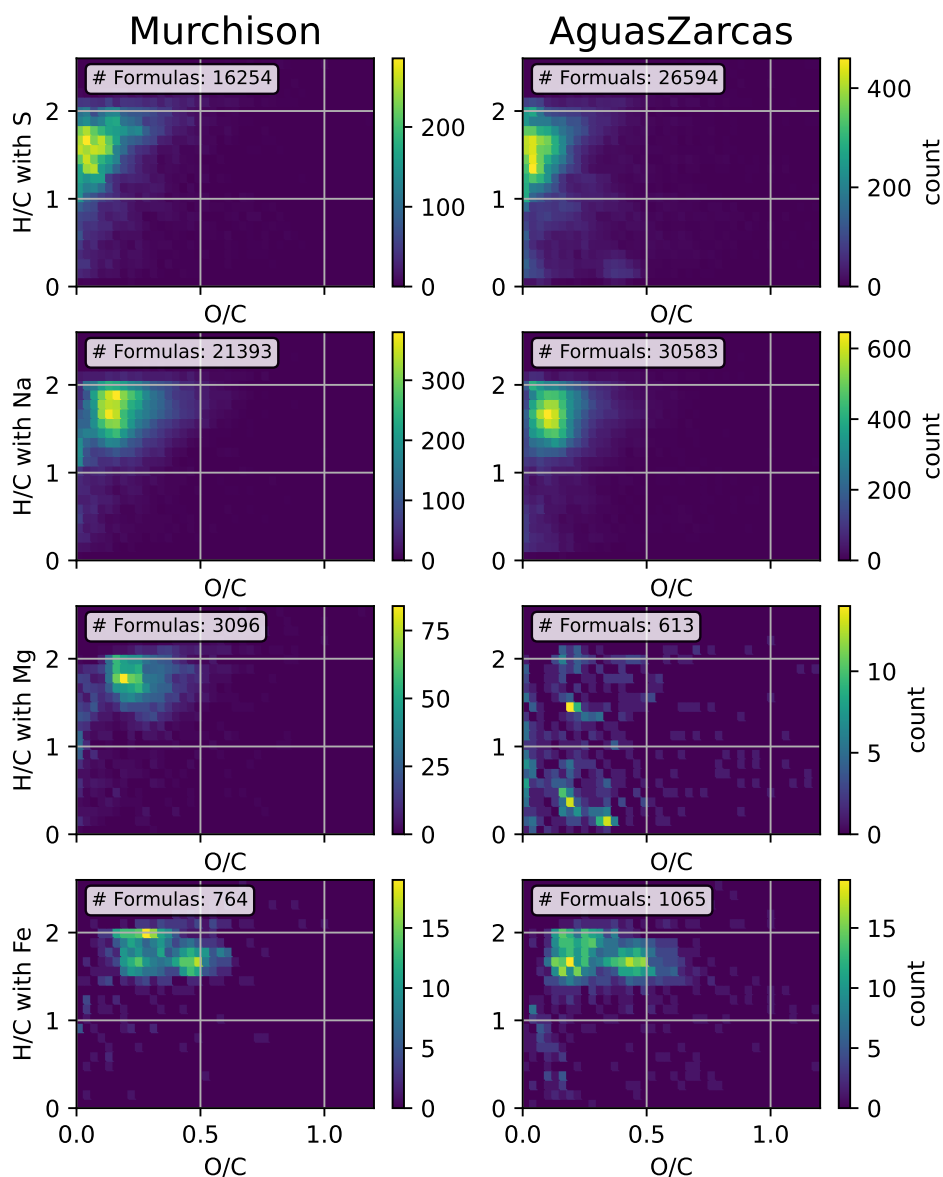


Figure B1. van Krevelen diagrams for isolated metal-containing compounds of all combined formulas found in Murchison versus Aguas Zarcas. The data behind these plots are available via doi:[10.5281/zenodo.18891055](https://doi.org/10.5281/zenodo.18891055).

strongly enriched in these two separate compositional domains. In contrast, Aguas Zarcas shows a reduced intensity of the $H/C \approx 2$ population and a shift of this aliphatic-like cluster toward lower H/C values (~ 0.8) at $O/C \approx 0.25$, indicating a comparatively more oxidized and/or less saturated Fe-associated organic pool, consistent with reported differences in alteration history and organic composition between CM chondrites.

These observations demonstrate that Fe does not randomly associate with the bulk organic matter but instead selectively binds to specific molecular classes, highlighting distinct metal-organic interaction regimes. Such behavior likely reflects interactions between organic matter and Fe-rich mineral phases (e.g., magnetite and Fe-bearing phyllosilicates) formed

during aqueous alteration on the meteorite parent body (A. E. Rubin et al. 2007; V. Vinogradoff et al. 2018). This coupling between inorganic and organic chemistry may play a key role in the preservation, transformation, and stabilization of complex organic matter in carbonaceous meteorites.

Table B1 showcases the first 10 entries of all formulas found for Murchison Metal Compounds with Mg, as of the data behind the chart in Figure B2 (B, left column scatter plots Murchison with Mg), all other data sets are available via Zenodo in a similar format in full length. Table B2 gives a summary for the number of all possible compound classes found for each meteorite sample as of the combined (+/−) ESI-MS data and also notes the remaining possible compound classes not been detected.

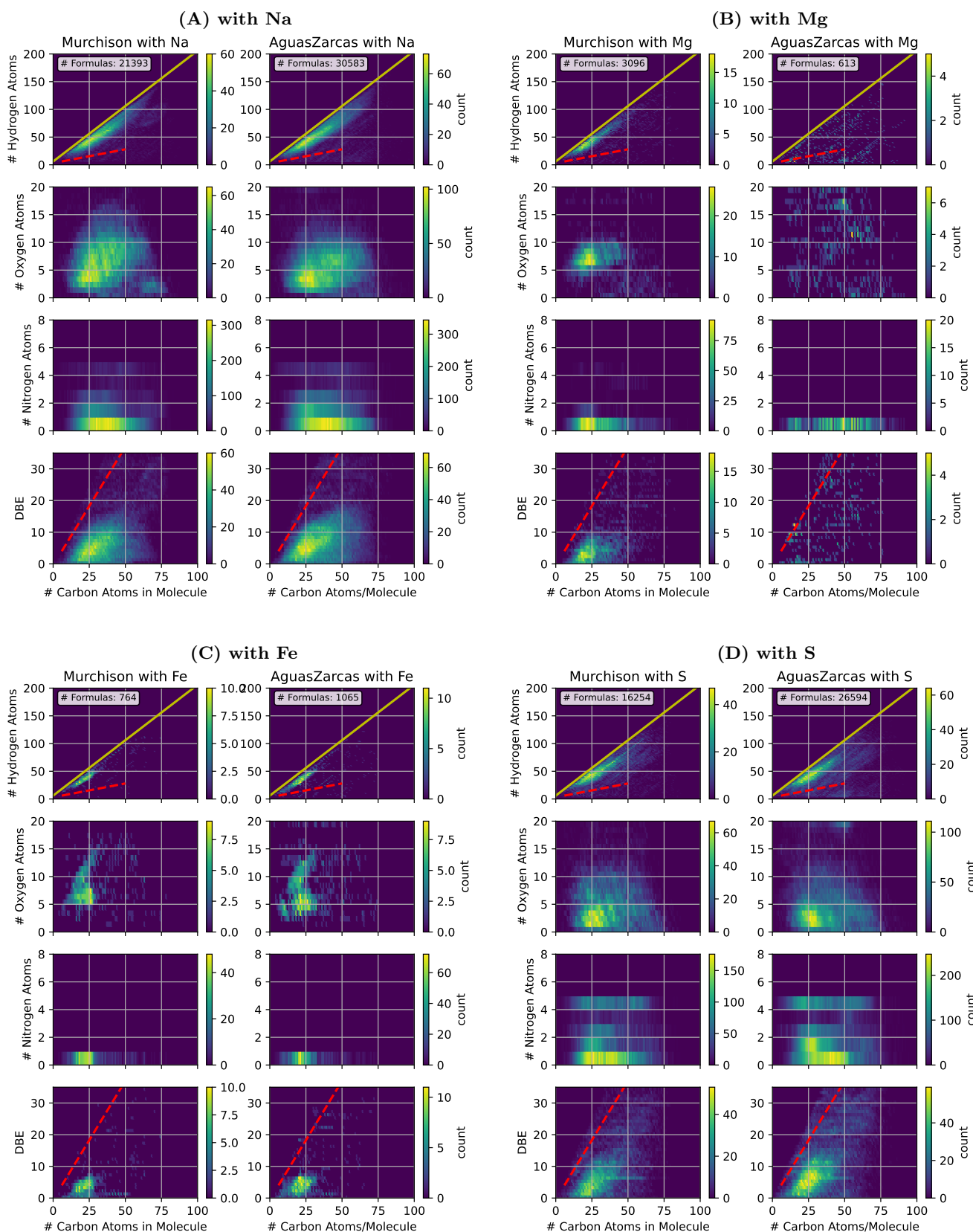


Figure B2. Isolated metal-containing compound compositions versus carbon number of all combined formulas found in Murchison versus Aguas Zarcas. Additionally, sulfur-containing compounds are shown. The data behind these plots are available via doi:[10.5281/zenodo.18891055](https://doi.org/10.5281/zenodo.18891055).

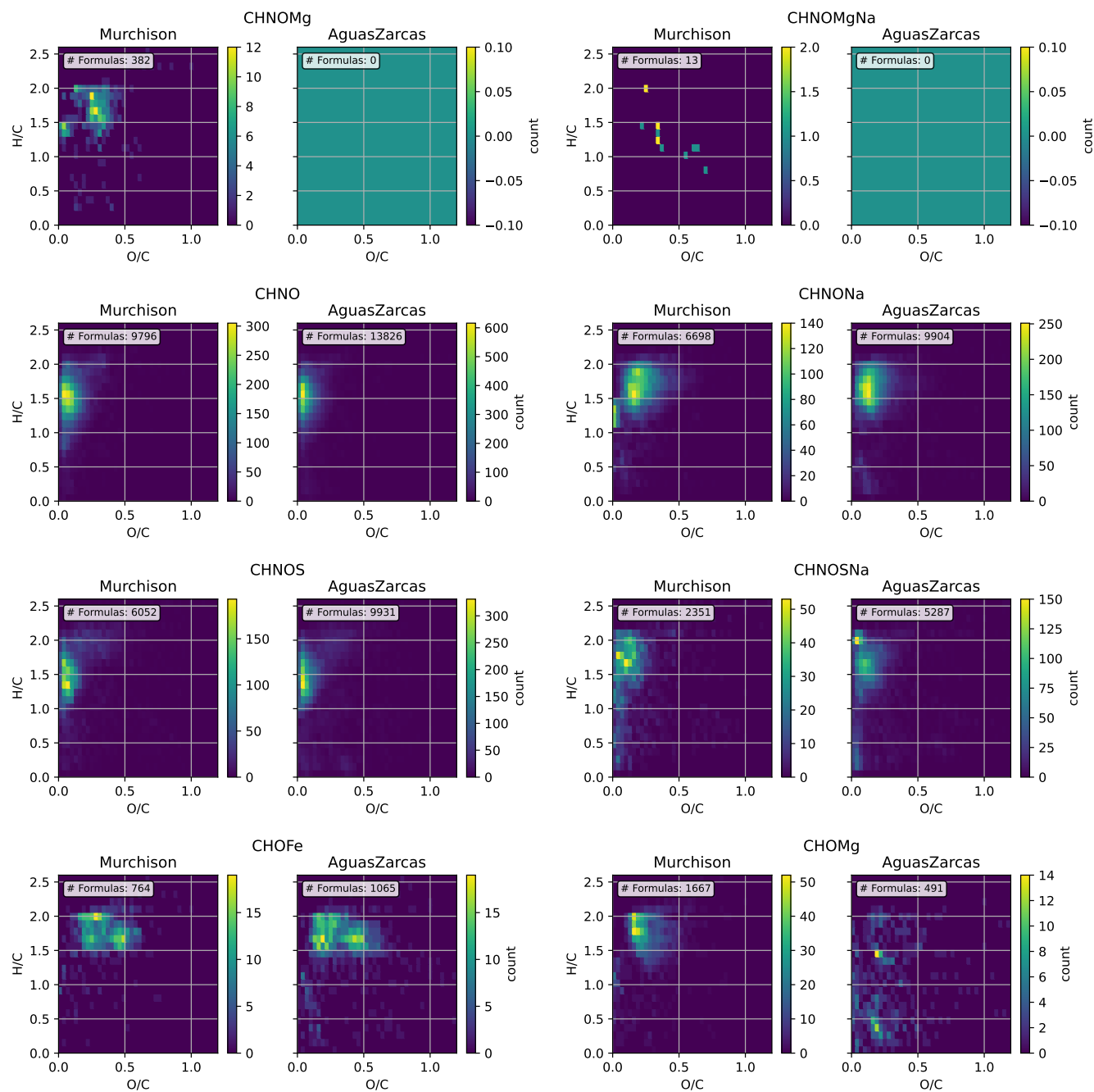


Figure B3. van Krevelen diagrams for all isolated formula compound classes with oxygen. The data behind these plots are available via doi:[10.5281/zenodo.18891055](https://doi.org/10.5281/zenodo.18891055).

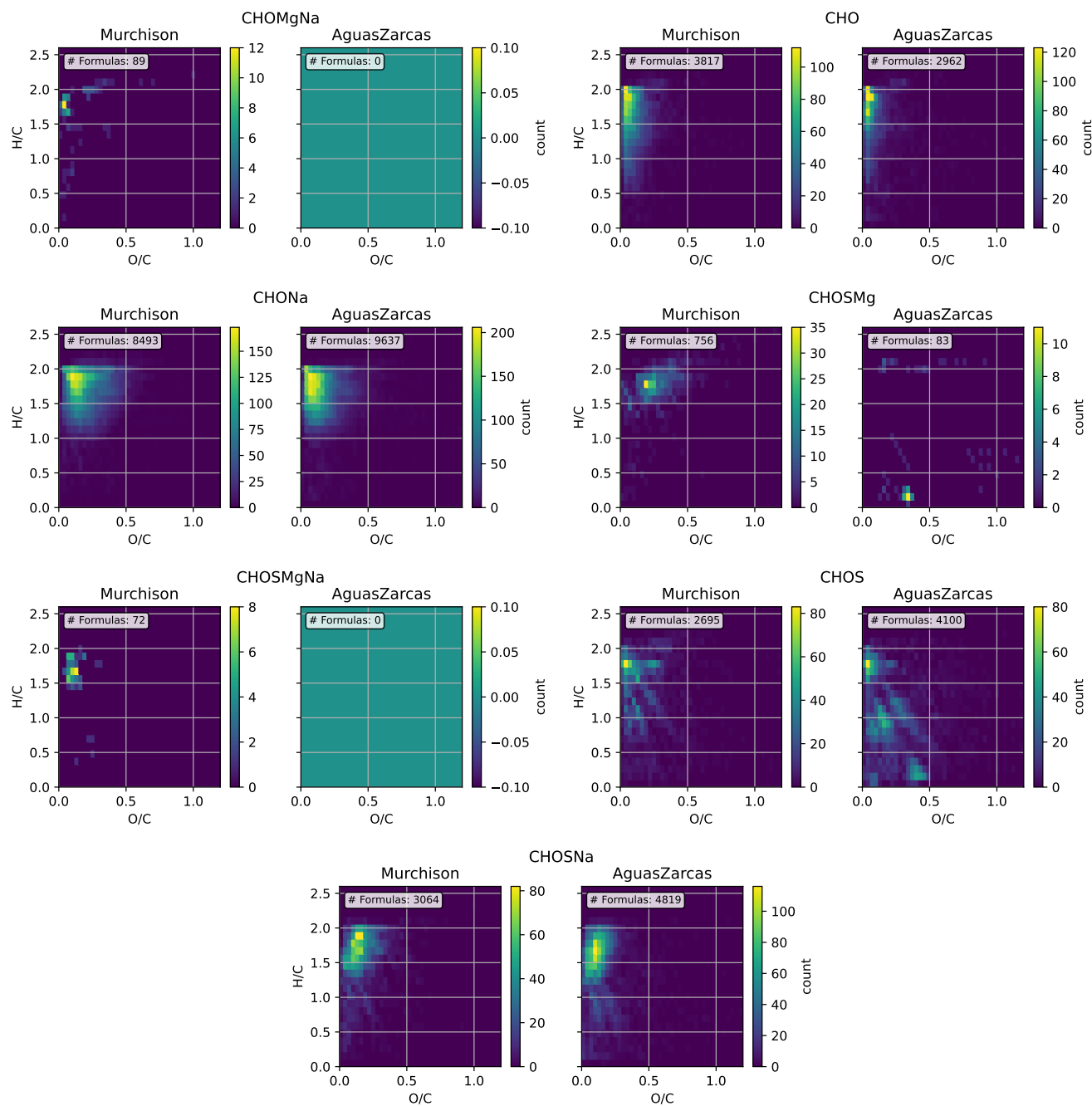


Figure B4. Continued: van Krevelen diagrams for all isolated formula compound classes with oxygen. The data behind these plots are available via doi:[10.5281/zenodo.18891055](https://doi.org/10.5281/zenodo.18891055).

Appendix C

Additional STM Overview Maps and AFM Images and Structures

Here, we provide additional STM and HR-AFM image data. Figure C1 shows two large-scale STM images of molecules on

a Cu(111) surface after flash deposition of Murchison material from the acetone extract, which preferentially contains more planar PAH-like species. For HR-AFM, the most suitable and planar candidates were selected (boxed) for further investigation.

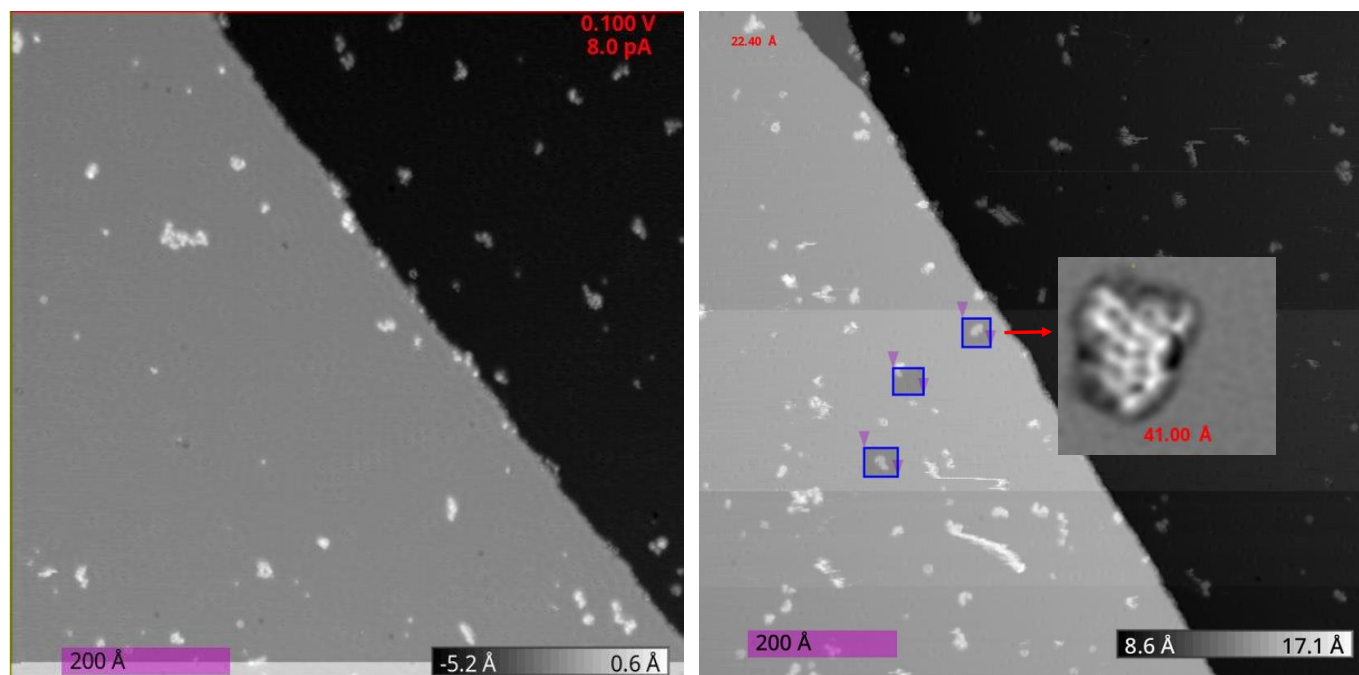


Figure C1. Supplemental STM overview images from the acetone extraction of Murchison meteorite. Both images show STM overview images of molecules as deposited on a Cu(111) surface. Images were obtained at 100 mV at 8 pA. A few actual imaged molecule “candidates” are marked with boxes within the right image, along with one HR-AFM zoomed-in molecular structure (presented in Figure 9, image 4).

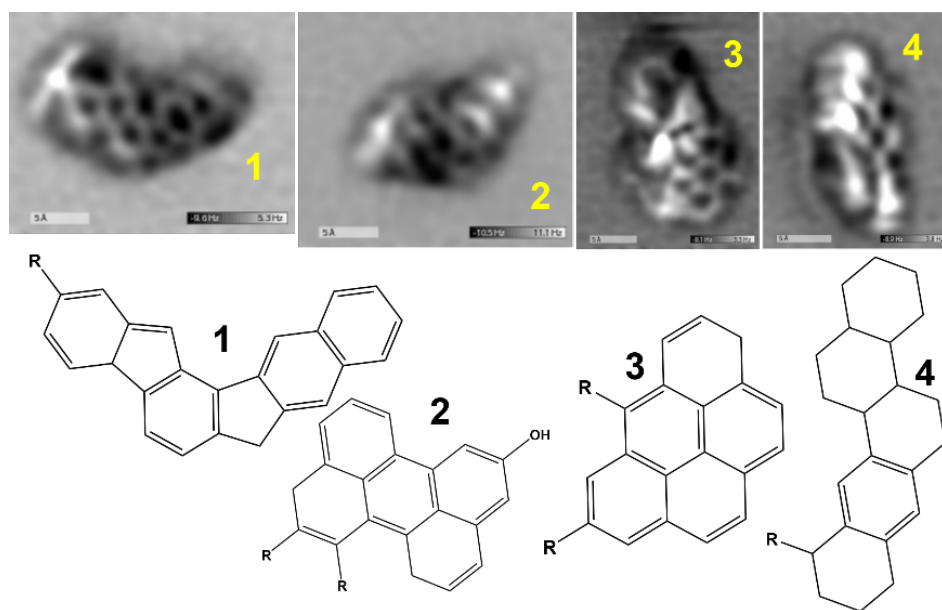


Figure C2. Supplemental HR-AFM images with proposed molecular structures from the acetone extract of the Murchison meteorite are shown. Four additional images are presented, containing structures similar to those discussed in the main article (Figure 10), obtained from the same extract. The light gray scale bar shown on the left in each image corresponds to 5 Å. The image contrast represents the frequency shift (typical range: -10 Hz, ..., $+5$ Hz), where brighter (more positive shift) indicates more repulsive tip-sample interactions, and darker (more negative shift) indicates more attractive interactions with the CO-functionalized tip. The $-R$ side groups may include $-H$, $-CH_3$, $-OH$, $-NH$, and $-NH_2$, among others. While the core structures are well resolved, uncertainty remains in the assignment of nonplanar side groups at the molecular edges, which often appear very bright due to their position partially out of the molecular plane. Very dark edge regions (closer to the substrate) are typically associated with radical sites (e.g., missing hydrogen atoms).

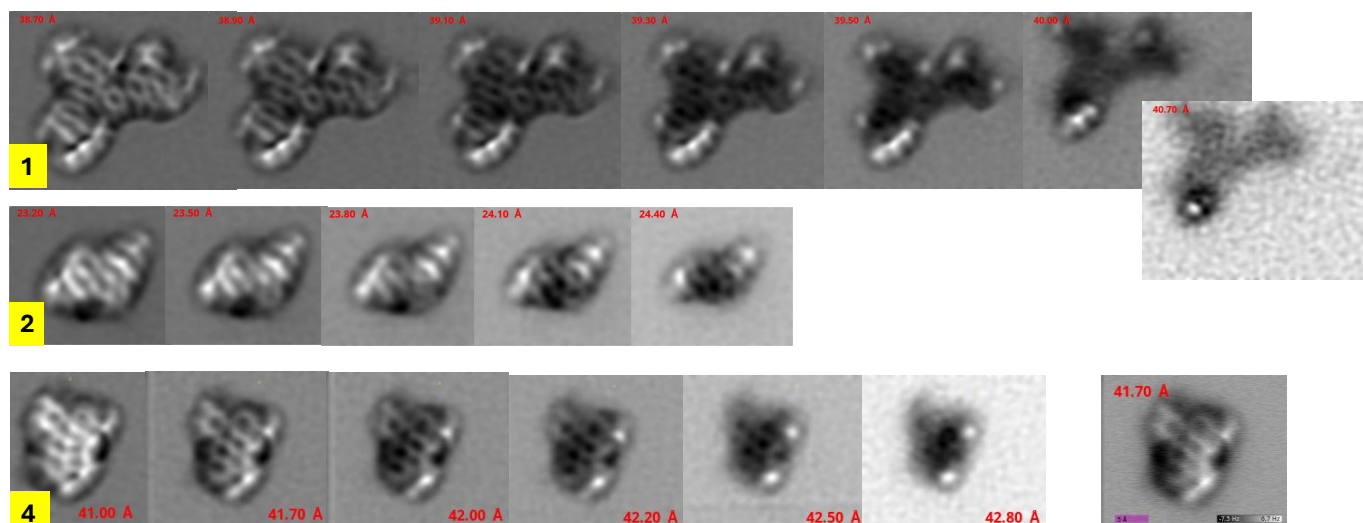


Figure C3. Supplemental AFM images detailing molecules labeled 1, 2, and 4 in Figure 10. We acquired sets of limited HR-AFM Z-slices for some molecules and present them here “as they are” for future advances in AI-based AFM image-to-structure techniques that may allow a better chemical structure evaluation.

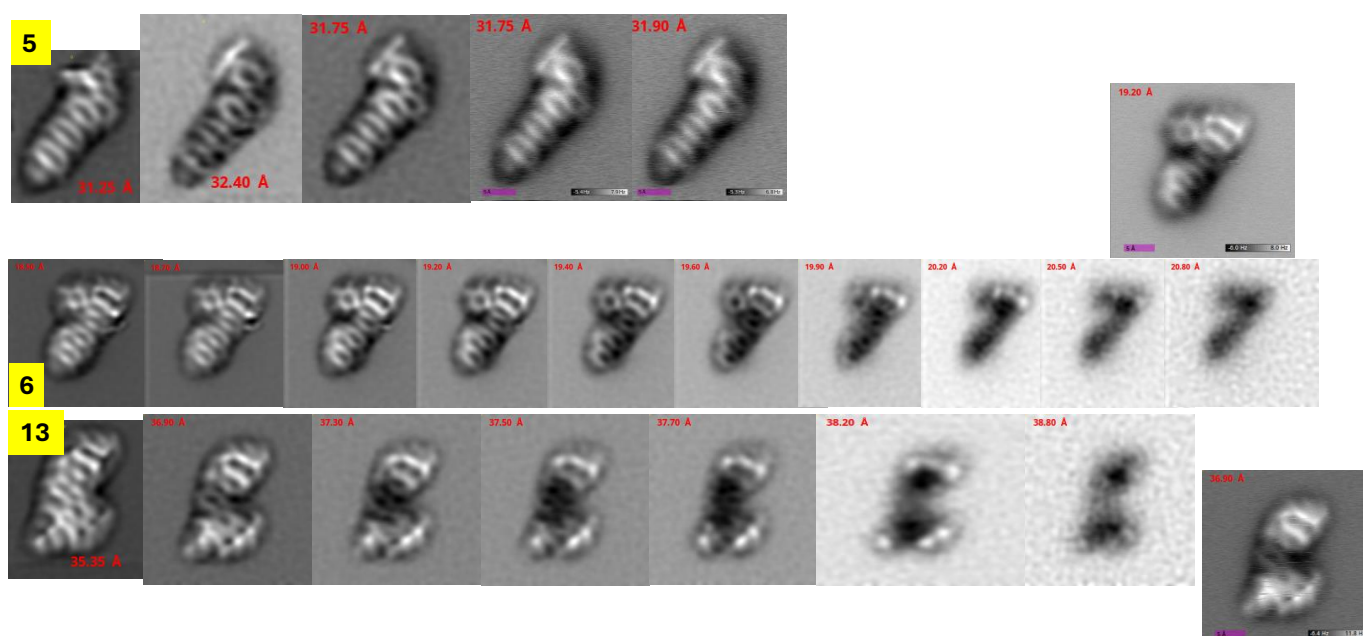


Figure C4. Supplemental AFM images detailing molecules labeled 5, 6, and 13 in Figure 10. We acquired sets of limited HR-AFM Z-slices for some molecules and present them here “as they are” for future advances in AI-based AFM image-to-structure techniques that may allow a better chemical structure evaluation.

Several challenges must be addressed for successful HR-AFM imaging, including molecular planarity, size and mobility on the surface, and possible interactions with the tip. A detailed discussion of these aspects is beyond the scope of this work.

Figure C2 extends the dataset presented in the main article (Figure 10). As a side note, the molecule labeled “1” in Figure C2 may contain a ring larger than C_6 (likely C_8) at the second position from the right. In the corresponding core model below, this position is indicated as a six-membered ring due to the current uncertainty.

Figures C3, C4, and C5 show force–distance slices of a few molecules presented previously (see molecule labels in yellow). These force–volume maps provide as detailed and complete a characterization as possible of the molecules by HR-AFM. The red numbers indicate the absolute tip position

(Z; not the distance, unknown offset) in ångströms; however, drift and creep effects must be considered.

Future advances in AI-based AFM image analysis are expected to improve the determination of the most probable chemical structures from experimental data for reasonably planar molecules on surfaces. At present, however, such methods are not yet sufficiently reliable or broadly available.

C.1. Abundances of Formulas from the Acetone Extract with Data from Related Actual Structures from HR-AFM Images

Alongside Figure 8 we also provide digital assets listing all molecule formulas from the chloroform extract (Table C1) and the acetone extract (Table C2), which can be found in the files `Formulas-CnHnNnOn-DBE-Chloroform.txt` and `Formulas-CnHnNnOn-DBE-Acetone.txt`, respectively, via doi:10.5281/zenodo.18891055 and are simple tables

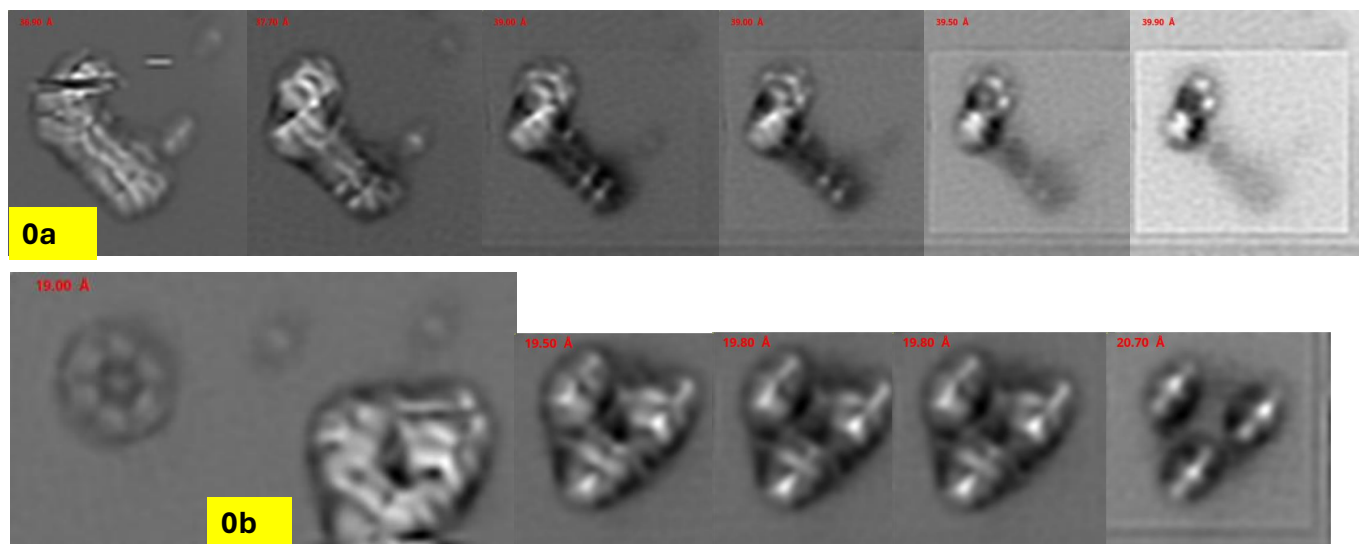


Figure C5. Supplemental AFM images not yet shown or discussed because they are hard to interpret with current knowledge. We acquired sets of limited HR-AFM Z-maps for some molecules and present them here. Future advances in AI-based AFM image-to-structure techniques may allow a better chemical structure evaluation.

Table C1

All Identified Formulas and DBEs of the Chloroform Extract

#C	#H	#N	#O	DBE
23	35	0	0	6
26	41	0	0	6
26	27	0	0	13
48	61	0	0	18
39	41	0	0	19
52	67	0	0	19
59	81	0	0	19

Note. This table is available in its entirety via doi:[10.5281/zenodo.18891055](https://doi.org/10.5281/zenodo.18891055). A portion is shown here for guidance regarding its form and content.

Table C2

All Identified Formulas and DBEs of the Acetone Extract

#C	#H	#N	#O	DBE
16	13	0	0	10
23	27	0	0	10
17	13	0	0	11
18	15	0	0	11
19	17	0	0	11
21	21	0	0	11
23	25	0	0	11
...				
20	27	4	20	9

Note. This table is available in its entirety via doi:[10.5281/zenodo.18891055](https://doi.org/10.5281/zenodo.18891055). A portion is shown here for guidance regarding its form and content.

containing rows with the C, H, N, O atom numbers as well as the calculated DBE.

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