

Chapter 2

Nitrogen in Solar System Minor Bodies: Delivery Pathways to Primeval Earth

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Abstract Oxygen isotope data point towards enstatite and ordinary chondrites as presumable building blocks of primordial Earth. Nitrogen was incorporated as nitriles to these first building blocks and was outgassed in the early stages of chemical segregation. However, giant impacts with planetesimals played an important role in partially eroding the atmosphere of Earth, and promoting thermal escape of diverse components. As a consequence, the Earth's atmospheric composition could have been subjected to important changes along the eons. A last, and probably less massive, delivery of volatiles took place at the time of a gigantic cataclysm known as Late Heavy Bombardment. During a short interval roughly between 3.9 and 3.8 Gyr ago, a gravitational migration inwards of Jupiter and Saturn occurred, that perturbed hundreds of small bodies rich in water, ammonia, methane and organic compounds that were stored until then in the outer regions. Current atmospheric signatures suggest that by that mechanism a continuous shower of outer-disk primordial components enriched the volatile inventory of terrestrial planets. The relevance of such contribution is still debated, but significant progress has been made in the last decades from the study of undifferentiated bodies. Consequently, planetary scattering of undifferentiated bodies delivered to Earth a significant fraction of minerals, and light elements that could have played a key role in the volatile enrichment the terrestrial crust. I suggest some unexplored pathways to allow a safe delivery of organics to Earth's surface, following recent evidence on meteoroid fragmentation, fireball spectra and Antarctic micrometeorite discoveries. Recent compositional studies of asteroids, comets and meteorites corroborate the need of having more precise data on the abundance and isotopic ratios of N in these minor bodies. Future space missions to primitive bodies like Rosetta, OSIRIS-

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Rex, Hayabusa II, or Marco Polo-R could help us to complete the big picture, and this chapter tries to compile our present knowledge of its delivery to Earth along the eons.

Introduction: The Sources of Terrestrial Nitrogen

Oxygen isotope ratios demonstrate a distinctive origin of meteorites in different parent bodies (Alexander et al. 2012). Such evidence points towards a differentiated accretion of planetary bodies from ring-like populations of building blocks mostly located at well-constrained solar distances, and then inheriting distinctive O isotope ratios. Oxygen isotope clues, and bulk chemistry point towards enstatite and ordinary chondrites as presumable building blocks of primordial Earth (Anders and Grevesse 1989; Wasson 2000; Lodders 2010). Such components were poor in volatile elements, but N was probably incorporated as nitrides (Rubin and Choi 2009). The primordial solar N and O isotopic composition has been inferred from first condensates (Meibom et al. 2007). This evidence allows the comparison of different sources of volatile elements, and helps establish plausible pathways for their delivery to Earth (Marty and Yokochi 2006). It seems likely there was an N-rich early atmosphere from direct outgassing. Despite this, during the late accretionary period large impacts eroded significant amounts of the Earth's volatile inventory. In this sense, a depletion in N and Xe has been noted with respect to other volatiles that are in chondritic proportions (Marty 2012). Such depletion could be explained as a consequence of large impacts producing a thermal escape and affecting the atmospheric components. In fact, a giant impact probably originated the Moon and eroded the early atmosphere of Earth (Cameron and Ward 1976).

Urey (1952) treated the origin of planets and their atmospheres in great detail for his time. Bukvic (1979) and Zahnle et al. (1988) and more recently Schaefer and Fegley (2007) and Schaefer et al. (2012) have modeled the equilibrium gas chemistry of an outgassed chondritic-vaporized reducing atmosphere, but several processes that can take place in the aftermath of the impacts are scarcely known (Trigo-Rodríguez and Martín-Torres 2013). Other authors have an opposite view where the outgassed vapors (Delano 2001), or the degassed vapors during impacts (Ahrens et al. 1989) produced oxidizing products ($\text{H}_2\text{O} + \text{CO}_2$). It is even possible that the degassed products varied significantly depending on variability of Earth's surface redox properties along the eons (Kasting and Catling 2003). Relative abundances of bioelements are in Table 2.1.

Current models accounting for the growth of terrestrial planets support the existence of a gigantic cataclysm over the entire solar system as the cause of the Late Heavy Bombardment (Gomes et al. 2005). Then, roughly between 3.9 and 3.8 Gyr ago, an inwards migration of Jupiter and Saturn occurred that gravitationally perturbed hundreds of minor bodies rich in water, ammonia, methane and organic compounds. That mechanism probably delivered to Earth a significant fraction of nitrogen and other bioelements currently present in the terrestrial hydrosphere, and

Table 2.1 Relative abundances of bioelements in the Sun, some Earth reservoirs and simple life forms (Adapted from [Golsmith and Owen 2003](#))

Element	Sun	Earth crust	Earth's atmosphere	Ocean	Microbium
H	91	0.14	< 0.001	11	63
O	0.08	46	21	86	26
C	0.03	0.02	0.04	0.003	6.4
N	0.010	0.002	78	5×10^{-5}	1.4
S	0.002	1.6	< 0.001	0.09	0.06
P	3×10^{-5}	0.11	< 0.001	6×10^{-6}	0.12

Table 2.2 Distinctive N isotopic composition of chondritic meteorites (According to data compiled by [Rubin and Choi \(2009\)](#))

Chondrite classes	$\delta^{15}\text{N}$	N location
Enstatite	≈ -20	Forming nitrides
Ordinary	-5 to 20	In chondrules
Carbonaceous	20 – 190	Mostly in matrix, and minor in chondrules

biosphere. Recent discoveries in minor bodies emphasize the enriching role of this late veneer population, and this chapter tries to compile current evidence in this regard. However, future sample-return missions like e.g. OSIRIS-Rex, Hayabusa II, or Marco Polo-R could provide a significant breakthrough in our knowledge ([Barucci et al. 2012](#)).

The D/H, and the $^{15}\text{N}/^{14}\text{N}$ Ratios in Transitional Bodies

The most primitive materials arriving to Earth come from undifferentiated comets and asteroids. A continuous flux of low-strength aggregates associated with comets are preferentially disrupted in the upper atmosphere ([Trigo-Rodríguez and Llorca 2006, 2007](#); [Trigo-Rodríguez and Blum 2009a](#)). Some arrive with the right geometry and velocity to survive as Interplanetary Dust Particles, but most are ablated and or disaggregated in the upper atmosphere as a consequence of their aerodynamic deceleration. Stardust studies of comet 81P/Wild 2 demonstrated that most of the N was present in the micron-sized matrix that was preferentially sputtered during aerogel capture in the tracks walls ([Brownlee et al. 2006](#)). In any case, in some surviving 81P/Wild 2 particles (e.g. shown by Febo) had matrix contained ^{15}N hotspots with isotopic signatures typical of the interstellar medium ([Matrajt et al. 2008](#)). This is not so surprising as carbonaceous chondrites also have distinctive heavy bulk N ($20 < \delta^{15}\text{N} < 190$) that are suggestive of a significant flux of interstellar materials moving inwards towards the disk regions where these primitive meteorites accreted. Consistent this picture and with the origin of the different chondrite classes, the amount of ^{15}N decreased going inwards in the disk (see [Table 2.2](#)).

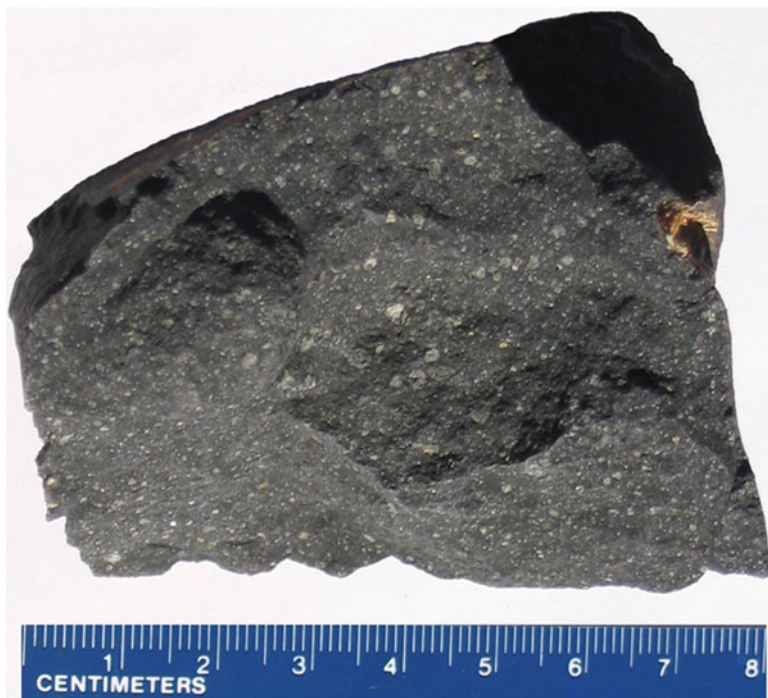


Fig. 2.1 A large specimen of the Murchison CM2 chondrite fell in Australia in 1969. The dark appearance of the matrix is due to its C content. *Grey* chondrules, and *white* Ca-Al rich inclusions (CAIs) are clearly distinguishable even at this naked-eye resolution (This picture made by the author is from a specimen belonging to the Leonard collection (IGPP/UCLA))

Chondritic meteorites are undifferentiated materials (e.g. non metamorphosed in large bodies) that comprise carbonaceous, ordinary and enstatite classes that are subdivided into 15 groups (Weisberg et al. 2006). All chondrites are considered chemically primitive by the fact that the ratios of their major, non-volatile elements (Fe, Si, Mg, Al, Ca, etc...) are close to those observed in the Sun (Anders and Grevesse 1989). It is thought that the different groups formed at different heliocentric distances, and slightly different times. Consequently, they inherited important chemical differences from the environment. Among chondrites, the carbonaceous chondrite (CC) class is thought to be especially pristine (Zolensky and McSween 1988). Inside CCs, several groups (mainly CI, CM, CO and CRs) exhibit important water contents and are formed by hydrated or aqueously-altered minerals (see e.g. Brearley 2006). Other volatile elements, e.g. N, are mainly located in the fine grained dust (so called *matrix*) that is cementing these rocks with typical grain sizes of 1 μm or less (see Fig. 2.1).

In summary, the different chondrite classes are basically conglomerates of fine dust (a mixture of silicates, oxides, metal, sulfides and organic constituents, see Fig. 2.2), chondrules, and refractory or mafic inclusions (Brearley and Jones 1998).

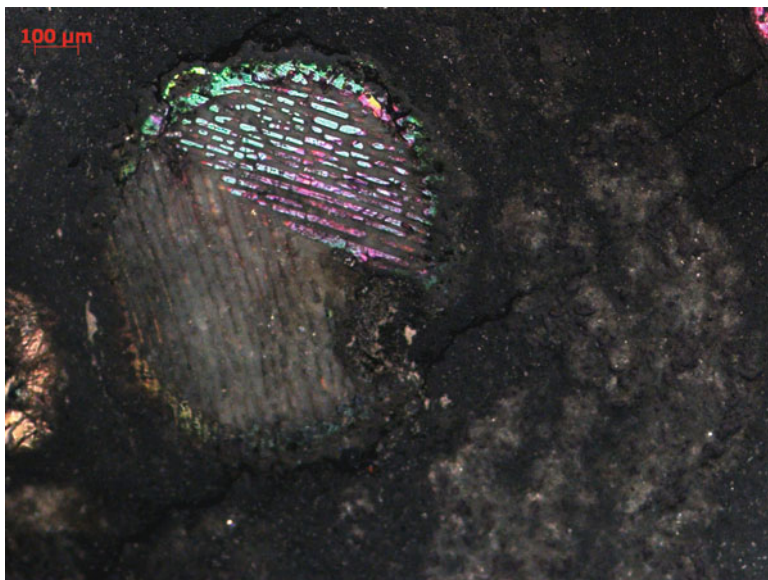


Fig. 2.2 A thin section of the Allende CV3 chondrite fell in Mexico in 1969. A barred olivine chondrule is seen in the *center*, and a CAI in the *bottom-right* quadrant. Notice the dark fine-grained matrix compacting the structure all around (J.M.Trigo/CSIC-IEEC)

The primordial water content in the different groups is difficult to assess, but those chondrites containing Fe-metal are indicative of being anhydrous in origin. We also know that many carbonaceous chondrite groups are unprocessed because they exhibit unequilibrated minerals, and also contain interstellar grains with isotopic anomalies that survived processing in the solar nebula and accretionary processes when incorporated to these rocks (Anders and Zinner 1993; Huss et al. 2006; Trigo-Rodríguez and Blum 2009b). Aqueous alteration plays against pristinity as there is clear evidence that some isotopic ratios (like e.g. D/H) are altered during parent body aqueous alteration processes (Alexander et al. 2012). Secondary minerals produced as consequence of aqueous alteration in carbonaceous chondrite parent bodies suggest that these processes occurred during the first 10 million of years after the birth of the solar system (Fujiya et al. 2013)

On the other hand, the N abundances and isotopic ratios in comets are poorly known. A significant achievement was obtained when the PIA mass spectrometer onboard Giotto (ESA) spacecraft was able to infer the composition of 1P/Halley dust particles. A big surprise was having most of the particles in the coma formed by light elements (H, C, N and O), the so-called CHON component (Kissel et al. 1986, see Fig. 2.3). Despite this, obtaining information about the light component is not trivial. For example, molecular nitrogen cannot be directly detected by remote spectroscopic techniques, and the usual inferences regarding N isotopic ratios are come directly from other species, such as NH_3 and HCN (Bockelée-Morvan

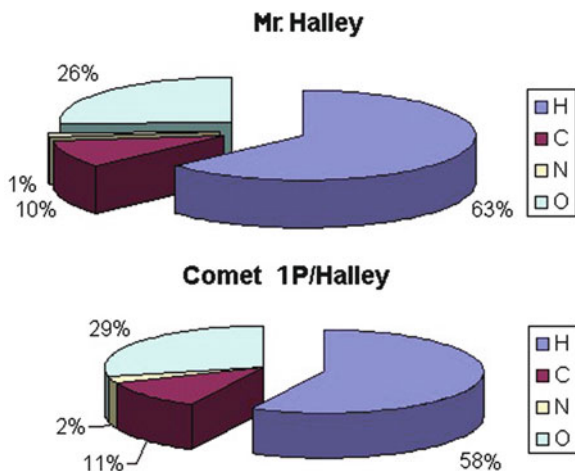


Fig. 2.3 A comparative view of the similitude between the proportions in the CHON elemental distribution found in comet 1P/Halley and humans

et al. 2004). All these species from which reasonable isotopic information is extracted are observed in cometary comae (see e.g. review by Rauer 2008). To have a better idea of the N content in comets we must wait for in situ studies of comets as precognized (Oró 1961). The Rosetta spacecraft is an ESA mission that plans to perform accurate studies of the isotopic composition of the CHON component in comet 67P/Churyumov-Gerasimenko by using *Ptolemy*: a gas chromatograph coupled to a mass spectrometer (Morse et al. 2008).

Calculations by Marty and Meibom (2007) suggest that a cometary contribution capable of fitting the abundances of noble gases is unable to produce more than $\sim 6\%$ of the nitrogen inventory in the terrestrial atmosphere. A similar amount is obtained in the review made by Marty (2012). Obviously we do not know the exact composition, and neither do we know the nature of the bodies that were able to reach the Earth during the LHB. Despite this, it is reasonable to consider the relevance of two still remaining water-rich populations. One is represented by the recently discovered Main-Belt comets (Hsieh and Jewitt 2006), and the other is represented by the Jupiter Family Comets that are collisionally generated fragments of Kuiper Belt Objects or KBOs (Jewitt 2008). In coming decades, we must explore those transitional objects that could be of interest for understanding this additional source of water and organics. An example is comet 29P/Schwassmann-Wachmann 1 that is considered the archetype of comets exhibiting outbursts and subsequent changes in their coma appearance and brightness (Sekanina 1982). This comet is defined as a Centaur due to its movement along a quasi-circular orbit that crosses the orbits of giant planets with eccentricity $e \sim 0.044$ and semimajor axis $a \sim 6$ AU. To get new clues on the structure, composition, and physical processes causing outbursts at these heliocentric distances we have promoted more than a decade of multiband photometric monitoring (Trigo-Rodríguez et al. 2008, 2010). During



Fig. 2.4 A thin section of GRA95229 from the NASA Antarctic collection that shows several silicate chondrules and the dark fine-grained matrix compacting the structure (J.M.Trigo/CSIC-IEEC)

an outburst its nuclear magnitude typically increases by 2 or 5 magnitudes in a behavior that seems unusual, but that would be typical for primitive objects like e.g. Centaurs, that evolve from Trans-Neptunian Objects (TNOs) located at the appropriated heliocentric distances.

Recent studies have identified a population of N-rich chondrites in the CR group of carbonaceous chondrites like e.g. GRA95229 (Fig. 2.4), suggesting that some regions in the protoplanetary disk had N-rich environments (Martins et al. 2007; Martins 2011; Pizzarello and Williams 2012). This enrichment in N and organics could be explained by gas drag effects that will transport inwards ice-rich bodies accreted in the trans-Neptunian region. A continuous disruption of such bodies at the particular time in which CR chondrites accreted could be envisioned as the source of N. In fact, high-eccentricity TNOs are probably the source of Jupiter Family comets (Emel'yanenko et al. 2004). Water and ammonia-rich TNO surfaces (covered by complex ice mixtures) have been found in the TNO population (DeMeo et al. 2010; Barucci et al. 2010). TNOs exhibiting such ices on their surfaces, could feasibly have experienced exhalation, perhaps by regular hydrothermal activity.

The contribution of the previously mentioned sources to the Earth's atmosphere and hydrosphere could be better explored when we acquire samples from primitive solar system objects. Future progress in modeling large impacts and the dynamic routes of minor bodies through solar system history could also be crucial (Kaula 1979; Wetherill 1985). For example, it has been recently demonstrated

that the cataclysmic impact forming the Moon probably did not completely erode the atmosphere of the Earth as previously thought (Genda and Abe 2003, 2005; Newman et al. 1999). This huge impact was probably an inflexion point followed by less massive impacts that brought volatile-rich materials and released reduced gases when subjected to subduction. Bukvic (1979) and more recently Schaefer and Fegley (2007) have modeled the equilibrium gas chemistry of an outgassed chondritic-vaporized reducing atmosphere, but several processes that can take place in the aftermath of the impacts are scarcely known. Other authors have an opposite view where the outgassed vapors (Delano 2001), or the degassed vapors during impacts (Ahrens et al. 1989) produced oxidizing products ($\text{H}_2\text{O} + \text{CO}_2$). Both scenarios are briefly explored, together with the influence of an enhanced shortwave solar activity during the early Hadean in Trigo-Rodríguez and Martín-Torres chapter (2013).

Ammonia, and Aqueous Alteration in Pristine Transitional Bodies

The chemical and structural differences observed in the different groups of carbonaceous chondrites are suggestive of their accretion in different outer regions of the protoplanetary disk where a moderate gas pressure and temperature allowed the ices and organics to remain in solid form. These aggregates formed the matrix in which the rest of components were accumulating. When the free-floating materials encountered each other under moderate relative velocities, progressive growth occurred until completion of the accretionary stage (Blum et al. 2006). Comets and carbonaceous asteroids were formed in the same way, but with different ice:dust ratios. As a direct consequence of their origin, the volatile elements were mostly retained in the fine-grained matrix.

Once the undifferentiated bodies completed their growing, some internal heat was released from the accreted radionuclides. Of particular interest in our discussion is the role of hydrostatic compaction in hydrous chondritic asteroids/comets. In other words, compaction due to the body's own gravity. In the last decade the idea of having a continuum of bodies with variable ice:rock ratio has received considerable support (see e.g. Gounelle 2011). In fact, accretionary models suggest that initial chondritic building blocks had porosities larger than 60 % (Blum et al. 2006; Trigo-Rodríguez and Blum 2009a). Depending on the ice:rock ratio and the final size of the body formed, they could have experienced hydrostatic inner pressures of the order of 0.1–3 MPa (Warren 2011). Under such hydrostatic pressure the water initially available as ice or as hydrated phases would be released, soaking the body. This type of aqueous alteration could be consistent with the observed typically static alteration features in hydrated groups like the CM chondrites (Trigo-Rodríguez et al. 2006, Rubin et al. 2007). Another clear type example are the Fe-rich aureoles found in the same group by Hanowski and Brearley (2000).

Significant progress has been made concerning the CM group of carbonaceous chondrites. The members of this group have experienced aqueous alteration to different degrees (Rubin et al. 2007). We infer that a large variety of mineral phases (e.g. phyllosilicates, sulfides, carbonates, oxides, and other poorly characterized phases or “PCP”) contained in these meteorites were produced via alteration of primitive materials by the pervasive action of water. The source of this water is unknown but could include water of hydration in phyllosilicates within the chondrites (Petaev and Wood 1998) or accreted ice (Stevenson and Lunine 1988). Parent-body processing like e.g. the produced primordially by radionuclide decay, or the induced by compaction of the materials due to gravity-sintering or impacts occurred along the eons caused water to be released from the initial host phases and produced aqueous alteration of primitive CM materials. Petrographic observations and the mineralogy of CM matrices are consistent with aqueous alteration at temperatures $< 400\text{K}$ (Bunch and Chang 1980). The presence of PCP clumps in CM matrices and the absence of glassy mesostases in CM chondrules attest to microscopic-scale aqueous alteration.

The presence of ice could have promoted hydro cryogenic alteration of anhydrous material through the action of unfrozen water over long time scales, but other processes involved soaking some regions of the CM parent body over shorter time scales. Obviously, an extended period of aqueous alteration would require the presence of liquid water and would imply a heating mechanism. As a consequence of heating, water exhalation or convection could be generated on the CM parent body (Young et al. 2003). The examination of newly discovered CM chondrites from Antarctica and hot desert regions provides new evidence bearing on the nature of aqueous alteration on the CM parent asteroid. We examined a suite of CM chondrites spanning the aqueous alteration sequence and found that one of these chondrites (MET 01070) contains cm-long, PCP-rich lenses that appears to be a product of aqueous flow on the parent body (Trigo-Rodríguez and Rubin 2006; Rubin et al. 2007). This is the first evidence of aqueous flow in CM chondrites (Fig. 2.5).

Discussion and Conclusions

N and other biogenic elements arrived at the Earth affected by different evolutionary cycles experienced in the inner solar system. The big picture is complex, but we are starting to understand the main evolutionary pathways. Current space missions are producing a large amount of information on the composition of comets and asteroids, which is paradoxical considering our scarce knowledge of the interaction of cometary materials with the atmosphere of Earth. However, Pierazzo and Chyba (2010) have recently made a significant computational progress developing a high-resolution hydrocode capable to predict shock temperatures and amino acids survival under particular entry conditions. The idea is not new, the delivery of organic compounds from comets was revisited in a couple of pioneering papers

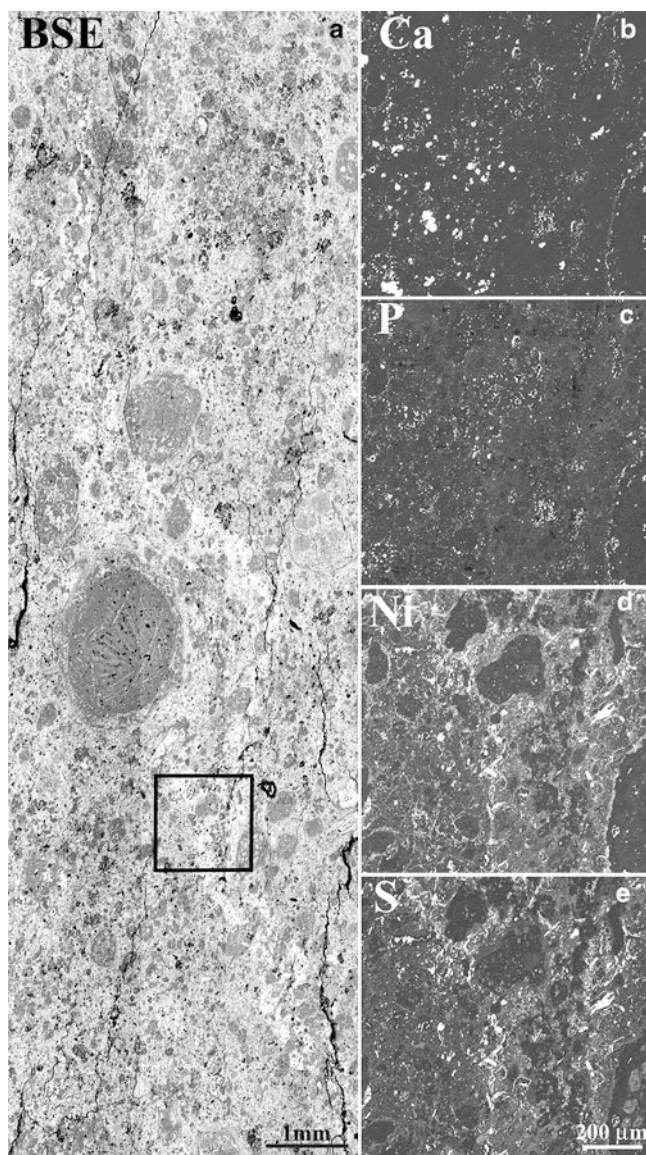


Fig. 2.5 A PCP lens identified in the SEM image of the Antarctic CM chondrite MET 01070. A X-ray mapping of the *black square* shows in the right images the presence of different elements mobilized by aqueous alteration (From [Rubin et al. 2007](#))

([Chyba et al. 1990](#); [Chyba and Sagan 1992](#)). Current models suggest that water-rich asteroids and comets impacting the atmosphere of Earth are delivering volatile elements during the bolide phase, but the amounts of surviving organic species

are scarce if the impact geometry and velocity are not favorable for an efficient deceleration and settling of the materials in the atmosphere (Blank et al. 2001). With this in mind, other direct pathways should be explored. For example, during the LHB period the scattered asteroids/comets should be fragile and could be efficiently disrupted during close approaches to terrestrial planets. If this is correct, the dense dust trails produced could efficiently deliver pristine materials with lower entry velocities. Fireball spectroscopy of cm- to mm-sized meteoroids suggests that catastrophic disruptions in the atmosphere can disperse dust far away from the shock wave frontal region where the bolide experiences higher temperatures (Trigo-Rodríguez et al. 2003; Trigo-Rodríguez and Blum 2006). Consequently, the exposure to heat is minimized and there is room for a percentage of body survival, at least as small fragments that are slowly setting down towards the surface. Such a scenario could be consistent with the recent discovery of ultracarbonaceous micrometeorites in Antarctica (Duprat et al. 2010).

Consequently, the main conclusions of this brief review are:

1. Current evidence suggests that N, and other light elements are abundant in undifferentiated bodies e.g. chondritic asteroids, comets, and TNOs. They are present in the fine-grained matrix forming these bodies, but probably also associated with aqueous alteration minerals.
2. Mobilization of light elements during aqueous alteration processes occurred at early times in the evolution of minor bodies. Such alteration processes provide a way to avoid outgassing of volatiles.
3. Due to the intrinsic difficulties associated with the remote detection of N and its isotopic ratio in minor bodies, the best way to proceed is by promoting future sample return missions like OSIRIS Rex, Hayabusa II or Marco Polo-R. Developing new technology to achieve a cryogenic return of cometary samples could also be the following step after them.
4. A significant part of Earth's atmosphere could have been delivered at the time of the Late Heavy Bombardment. However, a better knowledge of the volatile abundances in comets will allow us to revisit current models.
5. Fragmentation of meteoroids in the upper atmosphere of Earth could have provided a significant pathway for the delivery of the CHON component to primeval Earth.

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