

Noble gases in Dome C micrometeorites - An attempt to disentangle asteroidal and cometary sources

Bastian Baecker, Ulrich Ott, Mario Trieloff, Cécile Engrand, Jean Duprat

▶ To cite this version:

Bastian Baecker, Ulrich Ott, Mario Trieloff, Cécile Engrand, Jean Duprat. Noble gases in Dome C micrometeorites - An attempt to disentangle asteroidal and cometary sources. Icarus, 2022, 376, pp.114884. 10.1016/j.icarus.2022.114884. hal-03854561

HAL Id: hal-03854561 https://hal.science/hal-03854561

Submitted on 16 Nov 2022

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.



Distributed under a Creative Commons Attribution - NonCommercial - NoDerivatives 4.0 International License

1	Noble gases in Dome C micrometeorites -
2	an attempt to disentangle asteroidal and cometary sources
3 4	Bastian Baecker ^{1,2†} , Ulrich Ott ^{1,2,3*} , Mario Trieloff ² , Cécile Engrand ⁴ , and Jean Duprat ⁵
5 6 7 8 9 10	¹ Max-Planck Institut für Chemie, Hahn-Meitner-Weg 1, 55128 Mainz, Germany ² Klaus-Tschira-Labor für Kosmochemie, Institut für Geowissenschaften, Universität Heidelberg, Im Neuenheimer Feld 234-236, 69120 Heidelberg, Germany ³ MTA Atomki, Bem tér 18/C, 4026 Debrecen, Hungary ⁴ IJCLab, CNRS-Paris-Saclay, Orsay, France ⁵ IMPMC, CNRS-MNHN, Paris, France
11 12	*corresponding author; e-mail: uli.ott@mpic.de; phone: +49-6136-42609 [†] current address: Elbeallee 51, 14612 Falkensee, Germany
13 14	Abstract
15 16 17 18 20 21 22 23 24 25 26 27 28 29 30 31 32 33 34 35 36 37	We have performed a comprehensive noble gas study, including the isotopes of krypton and xenon, on a set of micrometeorites (MMs) collected from surface snow at Dome C (DC) on the Antarctic plateau. He and Ne are generally dominated by a solar component, with lower ⁴ He concentrations and ⁴ He/ ²⁰ Ne ratios in crystalline (Xtal) compared to fine-grained carbonaceous (FgC) MMs. Concentrations of (surface-correlated) solar wind (SW) He and Ne in FgC MMs are at the high end of what has been seen in earlier work, whereas the abundances of (volume-correlated) Kr and Xe are similar to what has been found in previous studies of MMs. In most samples, isotopic ratios for Kr and Xe are in the usual range of Q-Kr and -Xe (the Q component is the dominating component in primitive macroscopic meteorites) and air. When quantifiable, cosmic ray exposure (CRE) ages based on cosmogenic ²¹ Ne and ³ He, in combination with the Poynting-Robertson effect, are broadly consistent with an origin of the MMs from the asteroid belt. An exception is an Xtal MM, which exhibits a cosmogenic ²¹ Ne concentration in agreement with an origin from beyond Saturn, consistent with a possible cometary origin. In addition, data for trapped noble gases in three (out of ten analyzed) DC MMs provide hints that these may be related to a cometary source. One sample, a fragment of a FgC MM, is of particular interest. This fragment exhibits a Xe composition, although with large analytical uncertainties, deficient in the heavy isotopes ¹³⁴ Xe and ¹³⁶ Xe. This is similar to the Xe isotopic pattern, probably related to cometary ice, measured by Rosetta in the coma of comet 67P/Churyumov-Gerasimenko. The same MM also has an unusually high ³⁶ Ar/ ³⁸ Ar ratio, consistent with Rosetta's Ar measurement (in this case the latter having a large uncertainty). The other hints are for two MMs, of crystalline (Xtal) type, that show Ne similar to that found in laboratory analysis of refractory grains captured from comet 81P/Wild 2 by the Stardust mission. Additional
38 39	³ He, similar to what has been seen in some cluster interplanetary dust particles (cluster IDPs).

- *Keywords:* micrometeorites; noble gases; cosmic ray exposure; comets; asteroids

42 1. Introduction

The study of noble gases (and other volatiles) in micrometeorites is of interest for 43 various reasons. One concerns the origin and evolution of the atmospheres of Earth and other 44 inner solar system rocky planets, which is one of the outstanding questions in planetology. A 45 variety of processes contributing the volatiles H, C, N and noble gases have been considered 46 (e.g., Maurette et al., 2000; Trieloff et al., 2000; Pepin and Porcelli, 2002; Marty, 2012; 47 Alexander et al., 2012; Albarède et al., 2013; Vogt et al., 2019; and references therein), 48 including the possibility of accretion of volatile-rich materials by the forming planets. This 49 50 infalling material may have been in form of planetesimals and/or interplanetary dust grains, from the outer asteroid belt, as well as cometary material from the outer solar system (Pepin, 51 2006; Marty, 2012; Bradley et al., 2014). Recent models for the origin of Earth's atmosphere 52 favor the contribution of a few percent of volatile-rich material, similar to carbonaceous 53 chondrites, accounting for the bulk of the volatile inventory (Marty, 2012; Albarède et al., 54 55 2013), but the timing of such contribution is still a matter of debate. Some contribution from comets appears to be required based on recent analyses of volatiles in the coma of the Jupiter 56 family comet (JFC) 67P/Churyumov-Gerasimenko (Marty et al., 2016, 2017). 57

58 Generally, one might expect that the contribution to the geochemical budget of the planets arising from infalling dust and planetesimals might be similar in composition to that 59 from primitive meteorites. In fact, Marty (2012) proposes that the isotopic compositions of H, 60 N, Ne and Ar of Earth's reservoirs can be explained by mixing of solar and chondritic end-61 members, with cometary contributions limited to a few percent. This was recently revisited 62 with a model proposing an accretion of the Earth from enstatite-chondrite-like composition, 63 with the need of later delivery of volatiles from an exogeneous source to account for the 64 isotopic composition of the ocean water (Piani et al., 2020). However, "classical", i.e. 65 macroscopic chondritic meteorites (i.e., with sizes in the range centimeter to meters) provide 66 only a small contribution to the influx of extraterrestrial material to Earth, at least over the 67 recent geological time scale. Instead, the dominant source of extraterrestrial material accreted 68 by the Earth today is represented by micrometeorites (MMs) with sizes less than a mm (Love 69 and Brownlee, 1993; Cremonese et al., 2012; Carrilo-Sánchez et al., 2020, Rojas et al., 2021). 70 Such MMs can be recovered from polar ice and snow from Greenland and Antarctica 71 (Maurette et al., 1991; Maurette et al., 2000; Taylor et al., 2000; Duprat et al., 2007; Rochette 72 73 et al., 2008; Genge et al., 2018). Estimates for the present flux of micrometeorites before atmospheric entry are based on the study of hypervelocity impact craters on the Long 74 Duration Exposure Facility (LDEF) spacecraft. From these, Love and Brownlee (1993) 75 derived a total mass influx accreted by Earth of 40 (\pm 20) x 10⁶ kg per year, while a 76 reevaluation of the LDEF data by Cremonese et al. (2012) suggests lower total mass influx 77 78 rates, between 7.4 (\pm 1.0) and 4.2 (\pm 0.5) x 10⁶ kg per year, the value depending on the assumed source - asteroidal or cometary. Estimates based on space observations and/or 79 modelling have, among others, been published by Grün et al. (1985), Nesvorný et al. (2010) 80 and, most recently, Carrillo-Sánchez et al. (2020). The latter yields a value of 10.2×10^6 kg 81 per year, and also provides estimates for fluxes to Mars of 0.75 x 10⁶ kg/year (surprisingly 82 low in comparison to earlier estimates, e.g., Flynn, 1996) and for Venus of 11.3 x 10⁶ kg/year, 83 respectively. 84

During their high velocity impact at Earth's atmosphere during entry, a portion of 85 interplanetary particles is fully vaporized, while the remaining part survives as melted, partly 86 melted and unmelted particles. Using Lidar observations of the vertical flux of mesospheric 87 Na (originating from the vaporized part), Gardner et al. (2014) inferred an intermediate influx 88 value (see this paper also for further estimates given in the literature). The fraction of the flux 89 that survives as particles reaching Earth's surface has been measured in various collections, 90 mainly from Antarctica. Values of $\sim 1.6 \times 10^6$ kg were obtained from the amount of cosmic 91 spherules recovered from the South Pole Water well (Taylor et al., 2012) and Transantarctic 92 Mountains (Suttle and Folco, 2020), where the latter corresponds to an average over a $\sim 10^6$ 93 year timescale. A measurement of the recent flux was performed at Dome C, using the 94 95 collection from which the particles studied in this paper come from, by Rojas et al. (2021). The accurate control on the exposure parameter S (expressed in m² times year) allowed to 96 infer an annual flux of about 5 x 10⁶ kg, summing both melted (i.e. cosmic spherules) and un-97 melted (uMMs) particles (Rojas et al, 2021). 98

Carrillo-Sánchez et al. (2020) combined a chemical ablation model (CABMOD) with a
Zodiacal Cloud Model (Zody), which also allowed to obtain separate predictions for the
contributions from different sources: asteroidal, Jupiter-family comets (JFC) and Halley-type
comets (HTC). In case of Earth (and Mars), their model predicts, in agreement with previous
studies (Nesvorný et al., 2010), the dominant flux to be in form of unmelted MMs, and the
dominant source to be comets, not only for the most fine-grained dust, but also in the size
range of the MMs (~200 µm diameter) that we studied in this work.

106 While MMs were initially mainly related to primitive CM-chondrite-like material based on mineralogical, chemical and isotopic studies, more diversity has become obvious as more 107 micrometeorite collections were performed and extensively studied. A substantial fraction of 108 the micrometeorite population (10 to 17%; Taylor et al., 2012; Cordier and Folco, 2014) may 109 be similar to ordinary chondrites and related to asteroids (Genge, 2008; Cordier et al., 2011a; 110 Suavet et al., 2011; van Ginneken et al., 2012; Imae et al., 2013; Noguchi et al., 2015; Shyam 111 Prasad et al., 2018), while a smaller fraction (less than 6%) has been linked to differentiated 112 asteroids such as the Vestoids (Taylor et al., 2007; Cordier et al., 2011b, 2012). There is also a 113 specific family of MMs, the so-called ultra-carbonaceous micrometeorites (UCAMMs), that 114 has a probable cometary origin. These particles, which have extremely high concentrations of 115 carbon (Dartois et al., 2018), have been identified in collections from both Concordia Station 116 (Dome C; Duprat et al., 2010) and Dome Fuji Station (Nakamura et al., 2005; Noguchi et al., 117 2015), and a cometary origin is inferred from their large deuterium excesses (Duprat et al., 118 2010) and the high nitrogen content of their organic matter (Dartois et al., 2013; Yabuta et al., 119 2017). Apparently, the diversity of micrometeorites is large, and selection effects may play a 120 role in the assessment (Genge et al., 2020), since larger MMs are more easily found on Earth 121 and easier to study. In any case, in the size range of the MMs that we have studied (Table 1), 122 123 there should be a fair chance to encounter cometary (in particular, JFC) particles (Rojas et al, 124 2021).

Based on an early set of data, Maurette et al. (2000) evaluated the possible contribution of micrometeorites to neon, organics, carbon dioxide, nitrogen and water on Earth and they 127 concluded that early accretion of primitive micrometeorites may have played a major role in

- the formation of the terrestrial atmosphere and oceans. In a more recent evaluation, Vogt et al.
- 129 (2019) conclude that interplanetary particles (IDPs) and micrometeorites (MMs) may very
- 130 well be an important source of Earth's neon (both interior and atmospheric). However, a
- 131 crucial ingredient in these estimates is the pre-Archean micrometeorite flux that has to be
- extrapolated from the present-day flux with an uncertainty that is still a matter of debate.

Another aspect of noble gases in MMs is that they may possibly allow to distinguish 133 between asteroidal and cometary sources, as the relative contribution of each source to dust 134 entering the terrestrial atmosphere is still an open question. A difficulty, in this respect, is that 135 Ne and He, in (unmelted) small particles that travelled through interplanetary space, are 136 generally dominated by implanted solar wind components. This has been demonstrated in 137 138 numerous studies, as, in fact, most studies of noble gases in MMs have concentrated on He and/or Ne only (see summary in Wieler, 2002a). Still, sometimes small excesses of ²¹Ne of 139 cosmogenic origin have been observed, which allow, in combination with the Poynting-140 Robertson effect (Wyatt and Whipple, 1950) estimating a cosmic ray exposure age and 141 inferring the place of origin. Using this approach, Trappitsch and Leya (2013) have re-142 examined the data available at the time and found three MMs that may have come from 143 orbital distances far beyond the asteroidal belt, i.e. of possible cometary origin. This is, 144 provided all cosmic ray exposure happened while the MMs were travelling as small particles. 145 However, as the authors note, it is difficult to rule out that much or even most of the exposure 146 occurred in a parent body regolith instead. 147

148 Less ambiguous would be signatures of trapped noble gas components. In case of an IDP ("Manchanito") the isotopic composition of Ne has been found to resemble Q-Ne (where 149 Q is the major host phase of Ar, Kr and Xe – though not of Ne – in primitive meteorites). 150 Since cometary Ne measured in samples returned by the Stardust mission (Marty et al., 2008) 151 also resembles Q-Ne in composition, this has been taken as an indication for a cometary 152 origin of this particle (Ogliore et al., 2020). Compared to He and Ne, the situation may be 153 more favorable for the heavier noble gases, in particular Xe. There are clear differences 154 between the composition of the trapped (i.e., non-radiogenic, non-cosmogenic) noble gas 155 component in meteorites and the atmospheres of Earth and Mars, the solar wind and, 156 157 apparently, also cometary noble gas. Compared to solar abundance ratios, trapped noble gases in primitive meteorites (the "planetary" component) are characterized by an elemental 158 abundance pattern strongly favoring the heavy over the light noble gases. Similarly, in 159 comparison to terrestrial and Martian atmospheres, marked differences appear, in particular in 160 the Kr/Xe ratio and the isotopic composition of Xe (see, e.g., discussions in Ott and 161 Begemann, 1985; Pepin, 1991; Owen et al., 1992; Owen and Bar-Nun, 1993; Owen, 2008; 162 Ott, 2014; Ott et al., 2019). These differences cannot be explained by simple mixing of 163 "planetary" and solar noble gases, unless some additional processing is invoked (e.g., Hébrard 164 165 and Marty, 2014). Of particular interest here is the isotopic composition of Xe measured by the Rosetta spacecraft in the coma of JFC 67/P Churyumov-Gerasimenko (Marty et al., 2017), 166 which is characterized by a unique pattern exhibiting a spectacular relative deficit of the two 167 heaviest isotopes, ¹³⁴Xe and ¹³⁶Xe, by roughly 30 and 60 %, respectively, compared to 168 common components. Identification in a MM would be an indication that it originated from a 169

parent body similar to 67/P-CG, more telling than the presence of Q-Ne, which appears also 170

- in primitive meteorites. Evidence for cometary Xe on the Moon may have been seen by 171
- Bekaert et al. (2017) and Pernet-Fisher et al. (2020), but the situation is complicated by the 172
- presence of contaminating (terrestrial) air and solar wind Xe. Modeling the Xe isotopic 173 composition these authors measured in lunar anorthosites required, in addition to solar wind 174
- 175 and air, a contribution from a component deficient in the heavy isotopes of Xe, such as in
- cometary (coma) Xe. While the origin of this Xe is not clear, one possibility is that it was 176
- introduced by micrometeorite impacts. Still, it is important to realize that Rosetta and Stardust 177
- sampled different types of cometary reservoirs, in addition to sampling different comets. Xe 178 in the cometary coma measured by Rosetta corresponds to that carried in ices (maybe
- 180 primarily the CO₂ fraction; Rubin et al., 2018), while Q-Ne was measured in solids captured
- from the cometary coma in the aerogel targets of the Stardust mission. 181

Previous studies of MMs including Ar and/or Kr and Xe are rare, see Marty et al. (2002, 182 2005), Osawa and Nagao (2002a), Osawa et al. (2000, 2003a, b, 2010) and Okazaki et al. 183 (2015). Among these, Osawa et al. (2003a, 2010) focused on cosmic spherules where noble 184 gas abundances were close to blank level in most cases. For the isotopes of Kr and Xe, fairly 185 precise data were obtained (in addition to data for He, Ne, Ar) by Sarda et al. (1991) on a 186 single large (230 µg) micrometeorite. Other results for the isotopes of Kr and Xe are reported 187 in Osawa et al. (2000) and Osawa and Nagao (2002a, b), whose data, however, do not include 188 the isotopes lighter than ⁸²Kr and ¹²⁹Xe. More recently, a few micrometeorite analyses were 189 included within an integrated study on Xe in Interplanetary Dust Particles by Spring (2014), 190 using the Relax (Refrigerator Enhanced Laser Analyzer for Xenon) instrument at Manchester 191 University (UK) (Gilmour et al., 1994; Crowther et al., 2008). Further results for Xe isotopes 192 in a few large unmelted Antarctic Cap-Prudhomme micrometeorites (Maurette, 2006) were 193 194 reported in an abstract by Olinger et al. (2013). In a previous study, we reported a full set of noble gas data in micrometeorites from the Transantarctic Mountains (Cordier et al., 2018; 195 Baecker et al. 2018). These micrometeorites accumulated since about a million years ago 196 (Rochette et al., 2008; Folco et al., 2008) and were relatively large (several hundred µm). 197 Here we report new results on a set of recently fallen, i.e. recovered from surface snow, MMs 198 from the Concordia collection (Duprat et al., 2007), with sizes of typically ~200 µm. We also 199 include results for a single MM from Cap Prudhomme (Maurette et al., 1991; Kurat et al., 200 1994). 201

202

179

Experimental Procedures 2. 203

2.1. Dome C samples 204

Except for the one MM from Cap Prudhomme (Kurat et al., 1994), the analyzed 205 samples are part of the Concordia collection. They were collected at the Concordia station, a 206 station jointly constructed by the French and Italian Polar Institutes, which is located at Dome 207 C, about 3200 m above sea level on the Antarctic plateau, and about 1100 km from the coast 208 of Adélie land. For details about the collection protocol see Duprat et al. (2007). 209

- 210 Before allocation for noble gas analysis, the MMs had been stored under vacuum
- 211 conditions and had been handled in a clean room. During selection, the samples were
- observed under higher magnification (115 x) and sizes of the allocated specimens were
- 213 determined using a stereo-microscope with a μ m-scale as a reference. Roughly 1/3 of each (by
- volume) of a given MM was allocated for noble gas analysis at the Max-Planck-Institute for
- 215 Chemistry (MPIC) in Mainz, while the remaining sample was kept at IJCLab (Orsay). The
- 216 weights of the fragments for noble gas analysis were calculated using the approximate volume
- 217 (assuming sphere or cone-head shape) and a density of ~ 2.1 g/cm³, as appropriate for CM and
- CI chondrites (Britt and Consolmagno, 2003). After selection and size measurements, the
- samples were deposited between two glass slides and transported to the MPIC.

particle	fragment #*	size [µm]	weight [µg]	class ^{\$}
Dome C				
DC 06_07_213	PL 09_20 3 (1)	97 x 91	~ 1.1	Xtal
DC 06_08_01	PL06_09A 1	110 x 92	~ 1.4	Xtal
DC 06_09_11	PL 07_01A 11	137 x 111	~ 1.3	FgC/Sc
DC 06_09_50	PL 07_02B 2	141 x 134	~ 3.3	FgC
DC 06_09_57	PL07_02B 9 (1)	74 x 36	~ 0.15	FgC
DC 06_09_57	PL07_02B 9 (2)	19 x 18	~ 0.01	FgC
DC 06_09_63	PL07_02C 3	198 x 141	~ 3.3	FgC/Xtal
DC 06_09_141	PL07_04C 9 (1)	84 x 79	~ 0.7	Xtal
DC 06_09_149	PL07_05A 5 (1)	85 x 60	~ 0.3	Xtal
DC 06_09_189	PL07_06A 9	93 x 92	~ 1.0	FgC/Sc
Cap Prudhomme				
CP 9-1-1994	PL 10-109-D9	252 x 188	~ 6.2	Sc

221 <u>Table 1.</u> Dome C (DC) and Cap Prudhomme (CP) MMs analyzed for noble gases.

*Material available for noble gas analysis often consisted of several fragments. Where not the
 complete material was used, the fragment number is given in parentheses. The approximate

weight was calculated as described in the text.

 $^{\text{S}}$ Xtal = crystalline; FgC = fine-grained carbonaceous; Sc = scoriaceous.

226

Table 1 gives basic information about the analyzed samples. The respective sample 227 228 name for each Dome C sample consists of the following combination: DC (Dome C) 06 (year 2006) 09 (number of the nylon filter where the particle was selected from) 247 (number of 229 particle). Each particle was fragmented in a dedicated glass holder labelled, e.g., PL07 230 (year) 02B (glass holder) 7 (hole position). The Cap Prudhomme (CP) sample has a similar 231 232 code system. For the noble gas work, we concentrated on unmelted MMs since these are most likely to have retained original noble gas signatures. The classification in Table 1 follows 233 Kurat et al. (1994), Engrand and Maurette (1998) and Genge et al. (2008): FgC = fine-grained 234 carbonaceous, Xtal =crystalline (consisting of coarse-grained anhydrous minerals). Only the 235 CP sample is scoriaceous, with two additional DC samples (DC 06 09 11 and DC 236 237 06 09 189) partly scoriaceous.

238 2.2. Noble gas analysis

Details of our procedures for noble gas analysis of MMs at MPIC, Mainz, are given in 239 our reports on MMs from the Transantarctic Mountains (Baecker et al., 2018: Cordier et al. 240 2018) and in the PhD thesis of B. Baecker (Baecker, 2014). In short, in view of the expected 241 low gas amounts, a newly dedicated system was set up at MPIC that differs from our 242 conventional systems (e.g., Schwenzer et al., 2007; Cartwright et al., 2014) primarily by two 243 244 features: a) Laser heating for gas extraction in order to minimize extraction blank; and b) a high sensitivity Noblesse noble gas mass spectrometer (Nu Instruments) with multiple ion 245 counting (MIC; 8 channeltrons, plus a Faraday cup for larger signals) for simultaneous 246 isotope measurements, which maximizes the number of detected ions. This system requires 247

cross-calibration of the detectors and (minor) correction for cross talk, as described inBaecker et al. (2018).

Gas extraction was accomplished by heating with a 30 Watt CO₂ IR laser system (MIR 250 10-30, New Wave Research), after which the released gases were cleaned using successive 251 exposures to Zr-Al SAES getters. They were separated into He plus Ne, Ar, Kr and Xe 252 fractions, respectively, using temperature-controlled activated charcoal, and sequentially 253 admitted to the mass spectrometer for analysis. Full multi-ion collection (MIC) was used for 254 Kr and Xe only, where, for Xe, we used a three-step measurement routine (Baecker, 2014; 255 Baecker et al., 2018). In case of Ar, isotopes ³⁶Ar and ³⁸Ar were measured on the central 256 channeltron (IC3), while ⁴⁰Ar was measured using the Faraday cup. Similarly, ⁴He was mostly 257 measured by Faraday detector, while the ³He signal was detected on IC3. As for Ne, all 258 isotopes of Ne as well as mass 18 (water, used for correction at ²⁰Ne) were measured using IC 259 3. Masses 40 and 44, which were also used for interference correction, were measured by 260 MIC using IC0 and IC6. 261

The DC samples were measured following the measurement of the TAM MMs (Baecker et al., 2018), at which time extraction blanks had become lower and more stable. Including interfering non-noble gas contributions at the respective masses, they were, in cm³ STP, approximately as follows: ³He: $2x10^{-15}$, ⁴He: $6x10^{-12}$, ²⁰Ne: $4x10^{-13}$, ²²Ne: $7x10^{-14}$, ³⁶Ar: $4x10^{-13}$, ¹³, ⁴⁰Ar: $1.0x10^{-10}$, ⁸⁴Kr: $1x10^{-14}$ and ¹³²Xe: $3x10^{-15}$.

Interference corrections were applied at mass 3 for interference from not fully resolved HD/H₃, at the Ne isotopes for ${}^{1}\text{H}_{2}{}^{18}\text{O}$ as well as doubly charged ${}^{40}\text{Ar}$ and CO₂, and at the Ar isotopes ${}^{36}\text{Ar}$ and ${}^{38}\text{Ar}$ for HCl and hydrocarbons. These corrections were generally minor. Sensitivity and instrumental mass discrimination were determined by intermittent analyses of a noble gas mixture (obtained from *Air Liquide*) with atmospheric isotopic composition, to which extra ${}^{3}\text{He}$ had been added resulting in ${}^{3}\text{He}/{}^{4}\text{He} \sim 5x10^{-4}$. Errors in all the corrections are included in the reported (1 σ) uncertainties.

274

275 **3. Results**

276 **3.1. Helium and neon**

The results for He and Ne are reported in Tables 2 and 3. Similar as in previous 277 micrometeorite studies, the abundances of the major isotopes ⁴He and ²⁰Ne are dominated by 278 solar wind. No helium was detected in the two fragments of DC 06 09 57, while in two cases 279 (DC 06 09 141 and DC 06 09 189) we found ⁴He, but ³He was indistinguishable from 280 background. The data for those MMs, where both isotopes were detected, are displayed in 281 Fig. 1, a plot of ³He/⁴He in 10⁻⁴ units versus ⁴He in cm³ STP/g, alongside with the results from 282 previous studies and reference compositions (see figure caption for references). Separately 283 shown are the data from our previous analyses of the (larger) TAM MMs (Baecker et al., 284 285 2018; Cordier et al., 2018) and the (smaller) IDP Manzanito of Ogliore et al. (2020), which is of interest in the later discussion (chapter 4.5). 286

The isotopic ratios are broadly similar to previous studies, and also concentrations are 287 within the range previously observed. Again, as noted before, solar wind He is present not 288 only in unmelted MMs but also in such that have been partially molten (Sc = scoriaceous). 289 Interestingly, there is a clear distinction between Dome C MMs that are classified as Xtal 290 (crystalline) and others classified as FgC (= fine grained), FgC/Sc and FgC/Xtal. The highest 291 ⁴He concentrations are those of DC 06 09 189 (FgC/Sc) followed by DC 06 09 50 (FgC) 292 and DC 06 09 63 (FgC/Xtal), which are among the highest reported so far. Not surprisingly, 293 Xtal MMs with low ⁴He concentrations show high ³He/⁴He ratios, distinctly higher than in all 294 the trapped components shown, which is a clear indication for the presence of cosmogenic 295 ³He sited more retentively (volume-correlated) than solar wind He (surface-correlated). 296 Conversely, the more gas-rich samples show ³He/⁴He ratios similar to the trapped components 297 expected to be present, and it is difficult to assess the presence of cosmogenic He in these 298 cases. An exception may be FgC/Xtal DC 06 09 63, where the ratio is only slightly below 299 that of unfractionated trapped solar wind (see chapters 3.4 and 4.4). 300

The results for Ne are presented in Table 3. For the two fragments from DC 06 09 57, 301 we obtained only upper limits, otherwise amounts of 22 Ne for the DC MMs range between ~ 5 302 x 10⁻¹³ cm³ STP and ~4 x 10⁻¹² cm³ STP (concentrations between ~3 x 10⁻⁷ cm³ STP/g and ~7 303 x 10⁻⁶ cm³ STP/g). The scoriaceous MM from Cap Prudhomme is lower (2 x 10⁻¹³ cm³ STP 304 and 3 x 10⁻⁸ cm³ STP/g, resp.). Isotopic 20 Ne/ 22 Ne ratios are >11, which clearly indicates 305 substantial to dominant contributions from solar wind (SW and/or FSW). This is graphically 306 307 shown in the three-isotope plot of Fig. 2. Fig. 2a compares our DC and CP data with previous results for MMs (see Figure Caption for references). Like for most of the previous studies, the 308 data points plot roughly in the region between unfractionated solar wind (SW) and other 309 trapped components with lower 20 Ne/ 22 Ne (Ne-B, fractionated solar wind = FSW, Q, EA). 310 Contributions from cosmogenic Ne are small and difficult to establish precisely, if at all 311 (chapters 3.4 and 4.4). Again, this is typical for most MMs. Only in a few cases, in particular 312 for the scoriaceous MMs with low concentrations of solar Ne in our earlier TAM MM study 313 (Baecker et al., 2018; Cordier et al, 2018), a clear shift to higher ²¹Ne/²²Ne has been observed. 314

316 <u>Table 2.</u> He results for 10 different DC MM samples from 9 different MMs; as well as an additional

317 MM from Cap Prudhomme. Uncertainties in the last digits (1σ) are shown in parentheses and do not 318 include errors in sample weight. Where upper limits are listed, these are 2σ . For comparison, the

319 isotopic compositions of solar wind (SW), "fractionated solar wind" (FSW), Earth atmosphere (EA)

1519 Isotopic compositions of solar wind (SW), fractionated solar wind (FSW), Earth atmosphere (EA)

Sample	particle #		³ He	⁴ He	³ He	⁴ He	³ He/ ⁴ He
	purtien		(10 ⁻¹⁴ cm ³ STP)	$(10^{-12} \text{ cm}^3 \text{ STP})$	$(10^{-8} \text{ cm}^3 \text{ STP/g})$	(10 ⁻⁶ cm ³ STP/g)	(10-4)
DC 06_07_213	PL 09_20 3	3 (1)	1.3 (3)	16.2 (3.9)	1.22 (28)	15.2 (3.7)	7.96 (2.76)
DC 06_08_01	PL06_094	A 1	2.8 (3)	19.5 (3.9)	2.01 (27)	14.2 (3.0)	14.1 (3.3)
DC 06_09_11	PL 07_01A	A 11	2.8 (6)	86.7 (4.2)	2.13 (46)	67.1 (3.3)	3.18 (71)
DC 06_09_50	PL 07_02	B 2	363 (15)	12150 (188)	109.9 (4.6)	3682 (57)	2.98 (12)
DC 06_09_57	PL07_02B	9 (1)	< 0.5	< 10	< 3.6	< 64	
DC 06_09_57	PL07_02B	9 (2)		< 9		< 1141	
DC 06_09_63	PL07_020	C 3	131 (10)	3152 (57)	39.6 (2.9)	955 (17)	4.14 (30)
DC 06_09_141	PL07_04C	9 (1)		8.7 (4.3)		12.4 (6.1)	
DC 06_09_149	PL07_05A	5(1)		47.9 (4.3)		160 (14)	
DC 06_09_189	PL07_064	49	156 (3)	4799 (68)	156.3 (3.2)	4799 (68)	3.26 (9)
CP 9-1-1994	PL 10-109	-D9	< 3.2	8.86 (5.67)	< 0.52	1.4 (9)	
Component	References	³ He/ ⁴ He (10 ⁻⁴)	<u> </u>				
SW	[1]	4.645 (8	/				
"FSW"	[2], [3]	2.17 (5))				

and the primordial components Q (P1) and HL are listed at the bottom.

321 <u>References:</u> [1] Heber et al. (2012); [2] Benkert et al. (1993); [3] Wieler (2002a); [4] Busemann et al. (2000); [5] Ozima and Podosek

322 (2002); [6] Huss and Lewis (1994); <u>Note:</u> For more information regarding "FSW" see Wieler et al. (2007).

1.23 (2)

0.0140(1)

1.70 (10)

[4]

[5]

[6]

Q (P1)

EA

HL

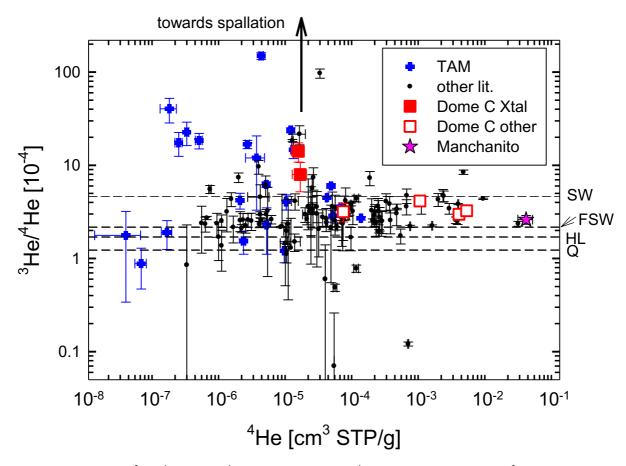


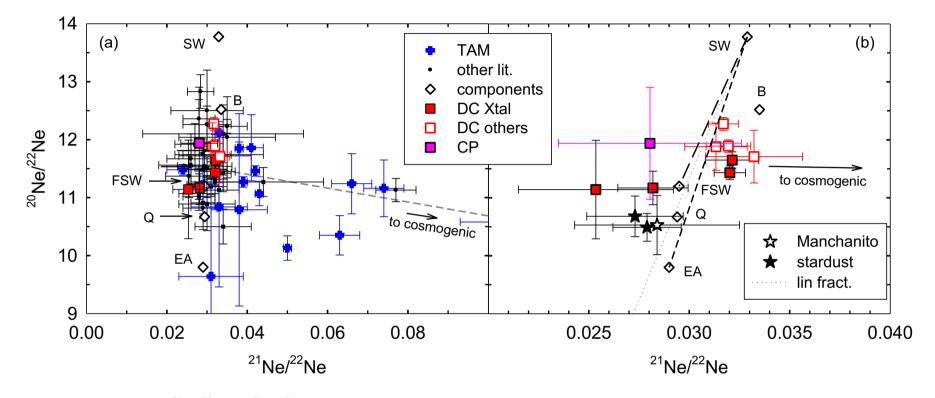
Figure 1. The ratio ³He/⁴He in 10⁻⁴ units is plotted vs. ⁴He concentration in cm³ STP/g for 324 Dome C micrometeorites. Error bars are 1_o. Data from our previous work on TAM MMs 325 (Baecker et al., 2018; Cordier et al., 2018) are shown for comparison, as well as data for other 326 MMs from the literature ("other lit."): Cap Prudhomme MMs extracted from ice, from Stuart 327 et al. (1999) and Marty et al. (2005); MMs extracted from snow at Dome Fuji (Osawa et al., 328 2000; Osawa and Nagao, 2002a; Okazaki et al., 2015); MMs extracted from ice near the 329 Yamato Mountains (Osawa and Nagao, 2002b; Osawa et al. 2003b); a large MM from 330 Greenland (Sarda et al., 1991); and the partially melted "cosmic spherule M240410 from 331 Osawa et al. (2010). Otherwise, data for cosmic spherules are not included. Note also that this 332 plot is somewhat biased since MMs with either ³He or ⁴He below detection limit are not 333 included. For reference the ³He/⁴He ratios of SW (Heber et al., 2012), "FSW" (Benkert et al., 334 1993; Wieler et al., 2007), Q (Busemann et al., 2000) and the HL component observed in 335 meteoritic nanodiamond residues (Huss and Lewis, 1994) are shown. Spallation He is 336 characterized by much higher ³He/⁴He (~0.2; e.g. Wieler, 2002b) than observed here, while 337 ${}^{3}\text{He}/{}^{4}\text{He}$ in the terrestrial atmosphere (1.4x10⁻⁶) is lower. 338

sample	particle #	²² Ne [10 ⁻¹² cm ³ STP]	²² Ne [10 ⁻⁸ cm ³ STP/g]	²⁰ Ne/ ²² Ne	²¹ Ne/ ²² Ne	³⁶ Ar [10 ⁻¹² cm ³ STP]	³⁶ Ar [10 ⁻⁸ cm ³ STP/g]	⁴⁰ Ar [10 ⁻⁶ cm ³ STP/g]	³⁸ Ar/ ³⁶ Ar	⁴⁰ Ar/ ³⁶ Ar
DC 06_07_213	PL 09_20 3 (1)	0.83 (3)	77.58 (7.76)	11.17 (29)	0.028 (2)	2.79 (34)	261 (32)	528 (65)	0.1841 (33)	202 (5)
DC 06_08_01	PL06_09A 1	3.22 (5)	234.9 (17.6)	11.43 (12)	0.032(1)	2.61 (32)	190 (23)	37 (21)	0.2051 (38)	19.7 (10.8)
DC 06_09_11	PL 07_01A 11	0.52 (3)	40.62 (3.82)	11.71 (45)	0.033 (2)	2.14 (27)	165 (21)	178 (27)	0.1906 (41)	107 (9)
DC 06_09_50	PL 07_02B 2	3.89 (6)	117.8 (4.0)	12.27 (11)	0.032(1)	19.7 (2.1)	596 (65)	181 (25)	0.1935 (23)	30.4 (2.6)
DC 06_09_57	PL07_02B 9 (1)	< 0.07	< 46.91			0.299 (132)	199 (88)	473 (22)	0.1726 (189)	237 (34)
DC 06_09_57	PL07_02B 9 (2)	< 0.07	< 771			2.98 (14)	37238 (1792)	76020 (4009)	0.1495 (39)	204 (4)
DC 06_09_63	PL07_02C 3	1.76 (10)	53.22 (3.56)	11.88 (41)	0.031 (2)	10.8 (4)	327 (14)	557 (30)	0.1830 (20)	170 (6)
DC 06_09_141	PL07_04C 9 (1)	0.22(2)	31.66 (5.69)	11.14 (85)	0.025 (4)	0.512 (132)	73.2 (18.8)	104 (31)	0.1731 (116)	142 (23)
DC 06_09_149	PL07_05A 5 (1)	2.06 (4)	688.0 (229.7)	11.65 (14)	0.032(1)	8.64 (21)	2881 (70)	162 (108)	0.1863 (36)	5.62 (3.76)
DC 06_09_189	PL07_06A 9	3.09 (5)	309.1 (31.4)	11.89 (11)	0.032(1)	14.6 (3)	1458 (31)	3177 (74)	0.1848 (36))	218 (2)
CP 9-1-1994	PL 10-109-D9	0.18 (3)	2.89 (52)	11.94 (97)	0.028 (5)	1.16 (5)	18.7 (8)	9.77 (72)	0.1719 (177)	52.1 (3.2)
component	references			²⁰ Ne/ ²² Ne	²¹ Ne/ ²² Ne				³⁸ Ar/ ³⁶ Ar	⁴⁰ Ar/ ³⁶ Ar
SW	[1]			13.777 (10)	0.03289(7)				0.183(1)	
"FSW"	[2, 3]			11.2 (2)	0.0295 (5)				0.205 (2)	
Q (P1)	[4]			10.67 (2)	0.0294 (10)				0.187(1)	
EA	[5, 6]			9.80	0.029				0.1885 (3)	298.56 (31
HL	[7]			8.50 (6)	0.036(1)				0.227 (3)	

340 <u>Table 3.</u> Ne and Ar results for 10 different DC MM samples from 9 different MMs as well as an additional MM from Cap Prudhomme. Uncertainties 341 in the last digits (1 σ) are given in parentheses. Where upper limits are given, these are 2σ . For comparison, the compositions of solar wind (SW), 342 "fractionated solar wind" (FSW), Earth atmosphere (EA) and the primordial components O (P1) and HL are listed at the bottom.

343 <u>References:</u> [1] Heber et al. (2012); [2] Benkert et al. (1993); [3] Wieler (2002a); [4] Busemann et al. (2000), the 20 Ne/ 22 Ne ratio is the mean for Q-Ne from Allende, Murchison,

Grosnaja and Chainpur; [5] Eberhardt et al. (1965); [6] Lee et al. (2006); [7] Huss and Lewis (1994); <u>Note:</u> For more information regarding "FSW" see Wieler et al. (2007).



347 <u>Figure 2:</u> Three isotope plots of 20 Ne/ 22 Ne vs. 21 Ne/ 22 Ne. Crystalline (Xtal) and other DC MMs are distinguished by different symbols. Error bars are 1 σ . 348 Plotted for reference are the trapped compositions SW (Heber et al., 2012), Ne-B (Black and Pepin, 1969), "FSW" (Benkert et al., 1993 and Wieler et al., 349 2007), Q (P1) (Busemann et al., 2000), Earth atmosphere (EA) (Eberhardt et al., 1965), and HL (Huss and Lewis, 1994).

a) For comparison, MM data from Dome Fuji (Osawa et al., 2000; Osawa and Nagao 2002a; Okazaki et al. 2015) and from the Yamato ice field (Osawa

and Nagao, 2002b; Osawa et al., 2003b) are included, as well as such from our earlier report on MMs from the Transantarctic Mountains (TAM: separate

symbols; Baecker et al., 2018). In order not to clutter the diagram, only literature data with an error of less than 0.5 in the 20 Ne/ 22 Ne ratio are shown. Some

353 scoriaceous TAM MMs fall outside the plotted range, shifted towards a cosmogenic composition.

- b) More detailed view of the DC MMs. Mixing lines SW-FSW and SW-EA, and the mass fractionation line (linear law) originating at the solar wind
- composition (Heber et al., 2012) are shown for reference, furthermore the composition measured for the IDP Manchanito (Ogliore et al., 2020) and the
 two more precise analyses of stardust track 41, aerogel cell C2044 (St-1 and Thera-2; Marty et al., 2008).

- Fig. 2b shows the range between ${}^{21}Ne/{}^{22}Ne = 0.02$ and 0.04 in more detail and concentrates on the samples of this study. Interestingly, as for He, there is a distinction between the MMs classified as Xtal and the others. The latter (i.e., the non-Xtal), together
- with two of the Xtal MMs, show higher 20 Ne/ 22 Ne than the other two crystalline MMs.
- 361 Compared to mixing lines between SW and FSW or EA, and also compared to compositions
- 362 for (linearly) mass fractionated SW, these MMs show a small shift towards elevated
- ²¹Ne/²²Ne, indicative of small cosmogenic contributions. On the other hand, Xtal DC
- 364 06_07_213 and Xtal 06_09_141 (as well as, with large error, the CP MM) plot to the left of
- 365 these lines. Their position is similar, although at somewhat higher 20 Ne/ 22 Ne, to what has been
- observed for the IDP Manchanito (Ogliore et al., 2020) and in the two more precise analyses
 of cometary Ne in stardust track 41, aerogel cell C2044 (Marty et al., 2008). The significance
- 368 of these observations will be discussed in chapter 4.5.

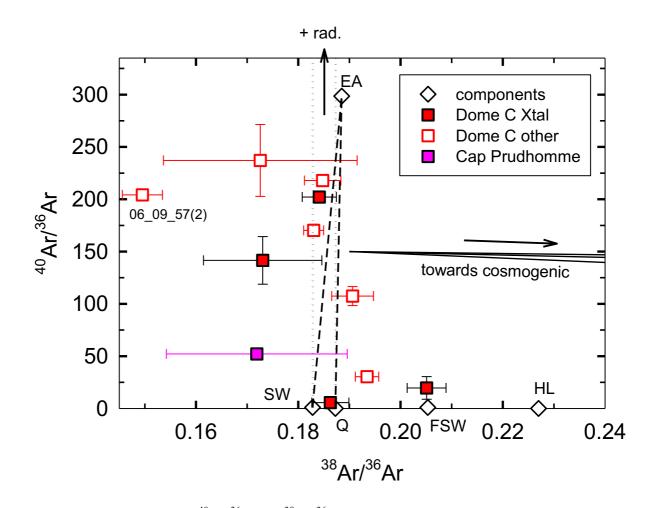
369 **3.2.** Argon

Results for Ar are given in Table 3 and depicted in a three-isotope plot of ⁴⁰Ar/³⁶Ar vs. 370 ³⁸Ar/³⁶Ar (Fig. 3). Except for the special case of DC 06 09 (see below), all of our MMs contain 371 substantial concentrations of 36 Ar (~7 x 10⁻⁷ to ~3 x 10⁻⁵ cm³ STP/g), along with 40 Ar/ 36 Ar 372 below the air ratio, thus clearly of extraterrestrial origin. Ratios of ⁴⁰Ar/³⁶Ar range from ~6 373 374 (Xtal DC 06 09 149) up to ~218 (Fg/Sc DC 06 189). The former is also characterized by the highest concentration of (extraterrestrial) ³⁶Ar, while the higher ratio for the latter may not be 375 surprising, since it is partly scoriaceous, and because scoriaceous MMs are more likely to be 376 contaminated by air Ar (Baecker et al., 2018). 377

Ratios of ²⁰Ne/³⁶Ar are in the range 2 to 5, with the exception of DC 06 08 01 (~14). 378 This is much lower than in the solar wind (~42; Heber et al., 2012; Vogel et al., 2009) and only 379 the latter value is similar to what we have previously estimated for FSW (~12; Baecker et al., 380 2018; based on data from Benkert et al., 1993). It appears likely, therefore that for Ar, besides 381 the solar wind contribution, there are also contributions from air and/or from primordial trapped 382 components such as Q. However, the implied extent depends very much on how strongly 383 fractionated the Ne/Ar ratio of the solar component is. This conclusion is borne out by the 384 position of the data points in Fig. 3, where several (with small errors) in ³⁸Ar/³⁶Ar plot close to 385 the composition of SW, perhaps slightly shifted towards air/Q. A few are further enhanced in 386 this ratio, in particular DC 06 08 01. Presence of cosmogenic ³⁸Ar would offer an explanation, 387 in principle, but this appears unlikely by comparison with the concentration of cosmogenic ²¹Ne 388 (chapter 3.4). Notably, ³⁸Ar/³⁶Ar for this MM happens to agree with that of pure FSW. 389

There are four cases where ³⁸Ar/³⁶Ar is lower than in SW, which has the lowest value 390 among the reference compositions that are shown. Three of them have very large uncertainties 391 and within 1σ overlap with SW, so we do not take this as significant. The fourth case is the 392 second specimen from DC 06 09 57 (labeled in Fig. 3), which we omitted in the discussion 393 above. This sample has by far the highest nominal concentration of 36 Ar of $\sim 3.7 \times 10^{-4} \text{ cm}^{-3}$ 394 STP/g – note that this has been the smallest analyzed sample by far and that uncertainties in 395 sample weight are not included in the errors given in Table 3. As it happens, the combination 396 of ³⁸Ar/³⁶Ar and ⁴⁰Ar/³⁶Ar could also result from mass-fractionated air Ar (with, possibly, a 397 small addition of radiogenic ⁴⁰Ar), but the mass fractionation would have to be enormous and 398

would have to favor the light isotopes, a very unusual situation. While one may also think of overcorrection for blank or isobaric interferences as a reason, this is also unlikely since the actual amount (rather than concentration) is not especially low in this sample. This is thus a surprising result, and we note that the same sample appears also to be unique in Xe, and that these two results might be related (chapter 4.5).



404

Figure 3: Three-isotope plot of ${}^{40}\text{Ar}/{}^{36}\text{Ar}$ vs. ${}^{38}\text{Ar}/{}^{36}\text{Ar}$ for, with 1σ error bars, separately for 405 crystalline (Xtal) and "other" MMs. Plotted for reference are the compositions of SW (Heber et al., 406 407 2012), "FSW" (Wieler, 2002a and Wieler et al., 2007), Q (P1) (Busemann et al., 2000), Earth atmosphere (EA) (Lee et al., 2006), and HL (Huss and Lewis, 1994). The dotted lines indicate the 408 position of data points with SW-like and Q-like ³⁸Ar/³⁶Ar and variable ⁴⁰Ar/³⁶Ar due to addition of 409 radiogenic ⁴⁰Ar, in order to guide the eve, while the dashed lines show the effects of mixing either 410 SW-Ar or Q-Ar with air. The solid lines trending to the right indicate the effect of mixing an 411 "arbitrary" (roughly in the center of the plot) composition with three types of cosmogenic Ar, 412 which, on the scale of the plot, are almost indistinguishable. From top to bottom these are: 413 cosmogenic Ar produced by GCR in MMs (Trappitsch and Leva, 2013); cosmogenic Ar as 414 typically found in meteorites (Wieler et al., 2002b); cosmogenic Ar produced in MMs by solar 415 416 cosmic rays characterized by a rigidity of 100 MV (Trappitsch and Leya, 2013). CM chondrite chemical composition was assumed for the production in MMs. Ar in the coma of comet 67/P 417 Churyumov-Gerasimenko has a highly uncertain ³⁸Ar/³⁶Ar ratio of 0.185±0.048 (Rubin et al., 418 2018), and a 40 Ar/ 36 Ar ratio has not been reported. 419

420

422 3.3. Krypton and Xenon

Tables 4 and 5 summarize the Kr and Xe results. Not listed are the rare isotopes ⁷⁸Kr, 423 ¹²⁴Xe and ¹²⁶Xe, for which no useful isotopic data were obtained. It is clear, though, that the 424 abundances of these isotopes are low. As a consequence, there are no significant cosmogenic 425 contributions that would make a noticeable effect on the ratios of the heavier, more abundant, 426 isotopes. Kr and Xe were not detectable or indistinguishable from blank in two MMs: Xtal DC 427 06 09 141 and the first (larger) of the DC 06 09 57 samples. Note that Spring (2014) also did 428 not find detectable Xe in their analysis of another portion of DC 06 09 57. Additionally, 429 krypton was not detectable in the scoriaceous Cap Prudhomme MM CP 9-1-1994. 430

Like in Ar, the second DC 06 09 57 sample is characterized by extremely high nominal 431 concentrations of Kr and Xe. While this is the smallest analyzed sample by far - with associated 432 large uncertainty in weight, which is not included in the nominal errors given in the tables - this 433 hardly offers an explanation for the much higher nominal concentrations, by roughly two orders 434 of magnitude in case of Kr (not counting DC 06-09 189, which is only ~3x lower). Otherwise, 435 Kr concentrations range between ~ 1 and $\sim 3 \times 10^{-8}$ cm³ STP/g (~ 1 to $\sim 7 \times 10^{-14}$ cm³ STP). 436 Measured amounts of ¹³²Xe range from ~1 x 10^{-15} cm³ STP to ~2 x 10^{-13} cm³ STP, with 437 concentrations mostly below 10⁻⁸ cm³ STP/g. The most notable exception, besides DC 06 189, 438 is, again, the second fragment of DC 06 09 57. This is, while the first fragment of this MM, 439 like the one analyzed by Spring (2014), was below detection limit. 440

Concerning isotopes of Kr, data scatter mostly around common trapped components (EA, 441 Q, SW), but SW is unlikely to make significant contributions to either Kr or Xe in most cases, 442 given the concentrations of the lighter elements and the relative abundances in the solar wind 443 (see chapter 4.3). Uncertainties in measured isotopic compositions are large compared to 444 diagnostic differences, anyway. The only possible exception is Xtal DC 06 07 213, which may 445 show a slight mass fractionation effect. While otherwise this sample does not stand out, its 446 position is similar in the three-isotope plots for Xe presented in Fig. 4, where the dotted and 447 dashed lines show the effect of mass fractionation starting with the composition of Q-Xe and 448 449 EA, respectively.

Isotopic ratios plotted in Fig. 4 are ¹³⁶Xe/¹³²Xe on the ordinate vs. ¹³⁴Xe/¹³²Xe (Fig. 4a) 450 and ¹³⁰Xe/¹³²Xe (Fig. 4b) on the abscissa. Xtal DC06 09 149 had the smallest measured 451 amount of Xe and its isotopic data have extremely large uncertainties. The nominal values plot 452 off scale. Reference compositions that are shown are SW, Q, EA (see Figure Caption for 453 references). Two mixing lines are shown in each figure: one (solid black line trending upwards) 454 455 between O and HL-Xe, the Xe carried by presolar nanodiamond, and another between O-Xe and Xe with the composition of cometary Xe (dashed dark cyan line), as inferred from 456 measurements in the coma of comet 67P/Churyumov-Gerasimenko by Marty et al. (2017). The 457 latter trends downwards. 458

459 <u>Table 4.</u> Kr results for DC micrometeorites ("DC") and a single MM from Cap Prudhomme ("CP"). No data are reported for 460 the rare ⁷⁸Kr isotope. Uncertainties in the last digits (1 σ) are given in parentheses, where the uncertainty in sample weight

461 is not included in the error of the concentration. For comparison, the compositions of solar wind (SW), Earth atmosphere

462	(EA) and the primordial compo	onents Q(P1) and HL are list	ed at the bottom.
-----	-------------------------------	------------------------------	-------------------

sample	particle #	⁸⁴ Kr [10 ⁻¹⁴ cm ³ STP]	⁸⁴ Kr [10 ⁻⁸ cm ³ STP/g]	⁸⁰ Kr/ ⁸⁴ Kr	⁸² Kr/ ⁸⁴ Kr	⁸³ Kr/ ⁸⁴ Kr	⁸⁶ Kr/ ⁸⁴ Kr
DC 06_07_213	PL 09_20 7 (3)	2.71 (60)	2.54 (56)	0.035 (5)	0.198 (17)	0.201 (12)	0.331 (20)
DC 06_08_01	PL06_09A 1	1.58 (60)	1.15 (43)	0.043 (8)	0.226 (27)	0.186 (18)	0.310 (27)
DC 06_09_11	PL 07_01A 11	1.18 (59)	0.91 (46)	0.038 (11)	0.274 (36)	0.209 (26)	0.312 (37)
DC 06_09_50	PL 07_02B 2	3.87 (61)	1.17 (19)	0.038 (4)	0.209 (12)	0.200 (9)	0.288 (13)
DC 06_09_57	PL07_02B 9 (1)	< 1.22	< 8.13				
DC 06_09_57	PL07_02B 9 (2)	2.06 (52)	258 (65)	0.053 (7)	0.214 (23)	0.201 (18)	0.324 (30)
DC 06_09_63	PL07_02C 3	6.86 (77)	2.08 (23)	0.045 (3)	0.215 (8)	0.206 (7)	0.287 (10)
DC 06_09_141	PL07_04C 9 (1)	< 1.04	< 1.49				
DC 06_09_149	PL07_05A 5 (1)	0.94 (51)	3.13 (1.71)	0.062 (13)	0.250 (38)	0.219 (27)	0.333 (44)
DC 06_09_189	PL07_06A 9	71.58 (3.19)	3.19 (38)	0.038 (1)	0.195 (7)	0.196 (6)	0.293 (8)
CP 9-1-1994	PL 10-109-D9	< 0.50	< 0.08				
component	references			⁸⁰ Kr/ ⁸⁴ Kr	⁸² Kr/ ⁸⁴ Kr	⁸³ Kr/ ⁸⁴ Kr	⁸⁶ Kr/ ⁸⁴ Kr
SW	[1]			0.0412 (2)	0.2054 (2)	0.2034 (2)	0.3012 (4)
Q (P1)	[2]			0.03937 (7)	0.2018 (2)	0.2018 (2)	0.3095 (5)
EA	[3]			0.039599 (20)	0.20217 (4)	0.2014 (2)	0.3052 (3)
HL	[4]			0.0305 (10)	≡ 0.1590	0.1989 (10)	0.3623 (18)

463 <u>References:</u> [1] Meshik et al. (2014); [2] Busemann et al. (2000); [3] Basford et al. (1973); [4] Huss and Lewis (1994) as re-calculated by

464 Busemann et al. (2000).

466 <u>Table 5.</u> Xe results for DC micrometeorites ("DC") and a single MM from Cap Prudhomme ("CP"). No data are reported for the rare isotopes 124 Xe

467 and ¹²⁶Xe. Uncertainties in the last digits (1 σ) are given in parentheses, where the uncertainty in sample weight is not included in the error of the 468 concentration. For comparison, the compositions of solar wind (SW), Earth atmosphere (EA) and the primordial components Q(P1) and HL are listed

469 at the bottom.

sample	particle #	¹³² Xe [10 ⁻¹⁴ cm ³]	¹³² Xe [10 ⁻⁸ cm ³ STP/g]	¹²⁸ Xe/ ¹³² Xe	¹²⁹ Xe/ ¹³² Xe	¹³⁰ Xe/ ¹³² Xe	¹³¹ Xe/ ¹³² Xe	¹³⁴ Xe/ ¹³² Xe	¹³⁶ Xe/ ¹³² Xe
DC 06_07_213	PL 09_20 7 (3)	0.91 (19)	0.85 (18)	0.060 (11)	0.947 (47)	0.135 (8)	0.841 (46)	0.412 (24)	0.394 (30)
DC 06_08_01	PL06_09A 1	0.39 (18)	0.28 (13)	0.041 (23)	0.921 (78)	0.137 (16)	0.632 (75)	0.431 (48)	0.342 (49)
DC 06_09_11	PL 07_01A 11	1.01 (19)	0.78 (15)	0.083 (10)	1.033 (44)	0.166 (9)	0.783 (40)	0.370 (21)	0.333 (24)
DC 06_09_50	PL 07_02B 2	2.87 (25)	0.87 (7)	0.072 (5)	0.995 (23)	0.162 (4)	0.784 (19)	0.397 (11)	0.349 (13)
DC 06_09_57	PL07_02B 9 (1)	< 0.17	< 1.12						
DC 06_09_57	PL07_02B 9 (2)	0.20 (8)	24.53 (10.21)	0.102 (64)	1.200 (139)	0.135 (48)	0.746 (157)	0.312 (78)	0.159 (91)
DC 06_09_63	PL07_02C 3	1.79 (26)	0.54 (8)	0.084 (8)	0.980 (31)	0.167 (6)	0.777 (29)	0.368 (16)	0.334 (18)
DC 06_09_141	PL07_04C 9 (1)	< 0.14	< 0.20						
DC 06_09_149	PL07_05A 5 (1)	0.11 (8)	3.53 (27)	0.028 (112)	0.911 (210)	0.124 (86)	0.898 (254)	0.384 (134)	0.513 (182)
DC 06_09_189	PL07_06A 9	17.27 (26)	17.27 (26)	0.073 (2)	0.991 (14)	0.153 (2)	0.782 (9)	0.386 (5)	0.318 (5)
CP 9-1-1994	PL 10-109-D9	0.90 (6)	0.14 (1)	0.077 (10)	1.035 (46)	0.158 (9)	0.869 (41)	0.362 (24)	0.299 (29)
component	references			¹²⁸ Xe/ ¹³² Xe	¹²⁹ Xe/ ¹³² Xe	¹³⁰ Xe/ ¹³² Xe	¹³¹ Xe/ ¹³² Xe	¹³⁴ Xe/ ¹³² Xe	¹³⁶ Xe/ ¹³² Xe
SW	[1]			0.0842 (2)	1.0405 (10)	0.1648 (3)	0.8256 (12)	0.3698 (6)	0.3003 (5)
Q (P1)	[2]			0.0822 (2)	1.042 (2)	0.1619 (3)	0.8185 (9)	0.3780 (11)	0.3164 (8)
EA	[3]			0.07136 (9)	0.9832 (12)	0.15136 (12)	0.7890 (11)	0.3879 (6)	0.3294 (4)
HL	[4]			0.0905 (6)	1.056 (2)	0.1542 (3)	0.8457 (13)	0.6356 (13)	≡ 0.6991

470 <u>References:</u> [1] Meshik et al. (2015); [2] Busemann et al. (2000); [3] Basford et al. (1973); [4] Huss and Lewis (1994) as re-calculated by Busemann et al. (2000)).

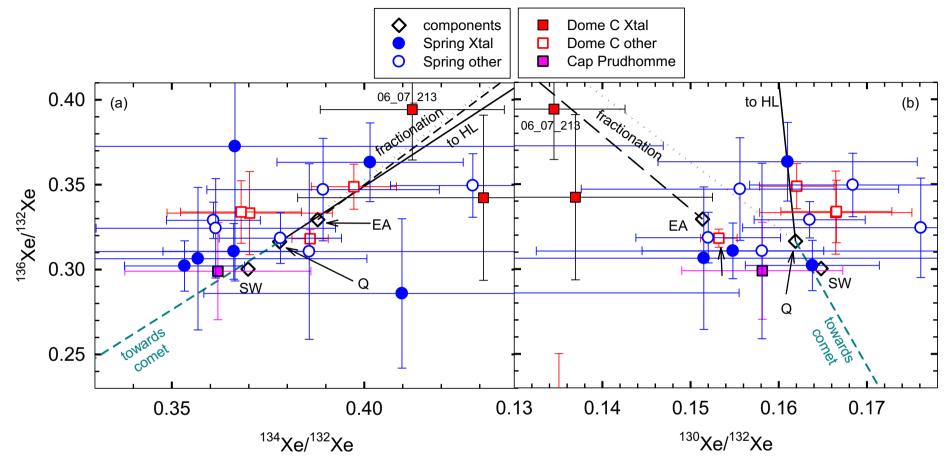


Figure 4. Three-isotope plots of 136 Xe/ 132 Xe vs. 134 Xe/ 132 Xe (a) and vs. 130 Xe/ 132 Xe (b). Data points for crystalline and other MM samples are shown separately. Error bars are 1 σ . DC 06_09_149 with very large uncertainties is not shown, while DC 06_09_57 (2) is off scale and discussed separately (chapter 4.5). Plotted for comparison are the compositions of air (Basford et al., 1973), Q-Xe (Busemann et al., 2000) and Xe-HL (Huss and Lewis, 1994; as recalculated by Busemann et al., 2000). Data from Spring (2014) obtained for MMs from Dome C and Cap Prudhomme are also included. The dotted and the dashed lines show the trend expected for mass fractionation starting from Q and air composition, respectively, while the solid line is the mixing line between Q and HL-Xe The dashed dark cyan lines are mixing lines between Q-Xe and Xe as reported for the coma of comet 67P/Churyumov-Gerasimenko by Marty et al. (2017).

Besides our own results, we have included in Fig. 4 data from Spring (2014), who 480 obtained data for Xe in the same MMs from Dome C analyzed here. Like our data, the Spring 481 data are distinguished in Fig. 4 between MMs classified as Xtal and others. Good agreement is 482 observed between her and the two data sets, but the uncertainties are large in both cases, due to 483 the small gas amounts analyzed. The Spring (2014) data do not include DC 06 09 57, since 484 485 Xe was not above extraction blank level. Again, this is in agreement with our result for the larger fragment we analyzed from this MM. The smaller fragment that we analyzed, however, 486 turned out different, having strange Ar isotopes (chapter 3.2), with very high abundances of Ar 487 and Kr, and comparably high Xe abundance. Concerning isotopes of Xe, this fragment is also 488 unique, with very low ¹³⁴Xe/¹³²Xe and ¹³⁶Xe/¹³²Xe. It plots off scale in Fig. 4, and we will return 489 to this observation below (chapter 4.5). 490

491 **3.4.** Cosmogenic noble gases

492 Contributions from cosmogenic noble gases in meteorites are commonly observed as excesses of ²¹Ne, ³He and, less frequently, ³⁸Ar. Such excesses are clearly visible only in few 493 of our MMs. This contrasts with the situation in the TAM MMs of Baecker et al. (2018), but 494 is similar to the majority of MMs analyzed in other previous studies (Dome Fuji and Yamato: 495 Osawa et al., 2000, 2003; Osawa and Nagao, 2002a, 2002b; Okazaki et al., 2015), the reason 496 being the high abundances of solar wind noble gases. For Ne, this is obvious in Fig. 2, where 497 Fig. 2a shows a comparison with the previous data, and Fig. 2b shows in more detail that our 498 data points plot close to trapped compositions that one might expect to be present: mixtures of 499 solar wind proper (SW; Heber et al., 2012) with fractionated solar wind (FSW; Benkert et al., 500 501 1993), Q (Ott, 2014) and Earth atmosphere, or simply mass fractionated solar wind (the dotted line). 502

Obviously, the data points to the left of the lines in Fig. 2b show no evidence of 503 cosmogenic Ne. Six others plot to the right, and we list in Table 6 the inferred concentrations 504 of cosmogenic ²¹Ne, together with the concentrations of trapped ²⁰Ne. We list two sets of 505 values obtained by using two different reference trapped compositions. However, as also 506 507 obvious in Fig. 2b, differences between the two approaches are small compared to the analytical errors. In one approach, following Baecker et al. (2018) to whom we refer for more 508 detail, the numbers were obtained from a 3-component deconvolution: a cosmogenic 509 510 endmember (with negligible influence of the exact choice), and two "trapped" endmembers, namely "fractionated solar wind" Ne (FSW; Benkert et al., 1993) and solar-wind Ne (SW; 511 Heber et al., 2012), as given in Table 3. The other approach, which was also used by, e.g., 512 Schmitz et al. (2019) in their study of fossil MMs, was simply considering the excess ²¹Ne 513 over the composition of (linearly) mass fractionated solar wind. Uncertainties of the reference 514 compositions were taken into account in calculating the listed uncertainties. Two additional 515 entries in Table 6 are upper limits for the two samples from DC 06 09 57, corresponding to 516 the actually measured amounts of ²¹Ne. 517

Table 6. Concentrations of cosmogenic ³He, ²¹Ne as well as "cosmogenic" ³⁸Ar and 519 concentration of trapped ²⁰Ne. Units are 10^{-8} cm³ STP/g, errors (1 σ) are in parentheses. The 520 cosmogenic abundances are sensitive to assumptions about the composition of the trapped 521 components, and two different sets have been used for both Ne and He, as described in the 522 text. In case of cosmogenic ²¹Ne, the upper values are those obtained from 3-component 523 resolution, while the lower values correspond to excesses over the linearly fitted SW mass 524 fractionation line. For cosmogenic ³He, the upper values are those obtained from an assumed 525 SW/FSW mixing ratio for the trapped component derived from the Ne data, while the lower 526 values assume a constant 3×10^{-4} for trapped ³He/⁴He. In case of DC 06:09 57 the listed upper 527 limits (2σ) correspond to total measured abundances instead. The case for the nominally high 528 abundance of cosmogenic ³He in DC 06 09 189 is marginal and listed in italics and small 529 print. Evidence for "cosmogenic" ³⁸Ar (calculated for the two cases with high ³⁸Ar/³⁶Ar in 530 Fig. 3, following Baecker et al., 2018) is also marginal and the values are listed in italics and 531 small print as well. 532

sample	particle #	²¹ Ne _{cos}	²⁰ Netr	³ He _{cos}	³⁸ Arcos
DC 06_07_213	PL 09_20 3 (1)		867 (90)	0.90 (24) 0.77 (24)	
DC 06_08_01	PL06_09A 1	0.53 (18) 0.41 (15)	2684 (51)	1.67 (24) 1.54 (24)	< 4.4
DC 06_09_11	PL 07_01A 11	0.13 (7) 0.11 (7)	475 (31)	0.30 (51) 0.12 (47)	
DC 06_09_50	PL 07_02B 2	0.09 (4) 0.06 (3)	1445 (25)	< 4.28 < 16.66	2.5 (2.2)
DC 06_09_57	PL07_02B 9 (1)	< 1.00	< 251	< 3.62	
DC 06_09_57	PL07_02B 9 (2)	< 12.66	< 5146		
DC 06_09_63	PL07_02C 3	0.05 (4) 0.03 (4)	632 (43)	12.48 (4.35) 10.92 (2.92)	
DC 06_09_141	PL07_04C 9 (1)		353 (47)		
DC 06_09_149	PL07_05A 5 (1)	1.44 (67) 1.13 (62)	8014 (188)		
DC 06_09_189	PL07_06A 9	0.49 (21) 0.36 (18)	3675 (73)	19.23 (6.75) 12.30 (10.40)	
CP 9-1-1994	PL 10-109-D9		35 (7)	< 0.45	

533

For cosmogenic ³He, except for two Xtal MMs (DC 06 07 213 and DC 06 08 01) 534 which clearly plot at higher ³He/⁴He than all trapped compositions (Fig. 1), results (or, for that 535 matter, the mere evidence for presence of cosmogenic ³He) depend sensitively on the choice 536 for the trapped ratio. Again, we list the results for two cases: in one case, again following 537 Baecker et al. (2018), we chose a composition for trapped ${}^{3}\text{He}/{}^{4}\text{He}$ assuming the same mixing 538 ratio (and nominal uncertainties) between SW and FSW for He as inferred for Ne. Since in 539 some cases, notably DC 06 09 63 and DC 06 09 189, the so obtained results appeared 540 unrealistic, and, since, in Fig. 1, ³He/⁴He for high (>10⁻⁴ cm³ STP/g) ⁴He concentrations 541 seems to level off at $\sim 3 \times 10^{-4}$, we used this value for the trapped component in the alternative 542 approach. An error of 0.2×10^{-4} in this ratio was propagated in this case. For the cosmogenic 543 component, we used ${}^{3}\text{He}/{}^{4}\text{He} = 0.20 \pm 0.05$ in both cases, but, as in Ne, the exact choice of the 544 cosmogenic composition has essentially no influence in the calculation of its abundance. With 545

the alternative assumption for the composition of the trapped component, the nominally high cosmogenic ³He abundance in DC 06_09_189 becomes a marginal case, but the abundance in DC 06_09_63 remains high. We note that the measured ratio in this MM is still below the ratio for unfractionated SW and so, in principle, the possibility remains that this cosmogenic ³He is not real. By comparison with other MMs it appears unlikely, however, to have such a high trapped ³He/⁴He ratio (see also chapter 4.4).

As for ³⁸Ar, there are two MMs that clearly show enhanced ³⁸Ar/³⁶Ar relative to SW, 552 "planetary" (Q) and air: DC 06 08 01 (Xtal) and DC 06 09 50 (FgC). The high ratios may, 553 in principle, be explained by addition of cosmogenic Ar. We have therefore calculated 554 nominal abundances of cosmogenic ³⁸Ar using the approach of Baecker et al. (2018), where 555 contributions to trapped Ar from SW, FSW and planetary Ar are estimated from the SW/FSW 556 557 mixing ratio at Ne and the Ne/Ar ratio of the different components. The values are listed in Table 6, but obviously, cosmogenic contributions are difficult to ascertain even in these two 558 cases. We will not consider cosmogenic ³⁸Ar in chapter 4.4. 559

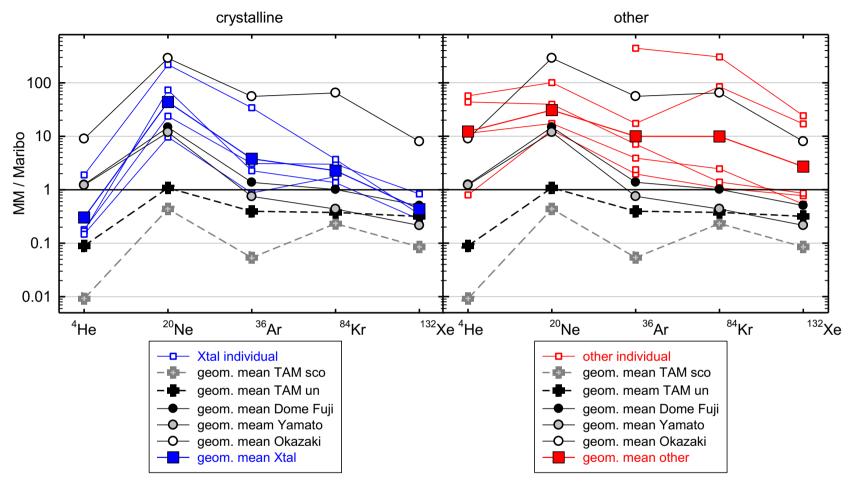
560

561 **4. Discussion**

562 4.1. Noble gas general pattern

We will concentrate on the MMs from Dome C in this discussion. The single particle from Cap Prudhomme, although being of larger size, is scoriaceous rather than primitive and has low noble gas abundances and, therefore, does not add information.

As common in meteorites with implanted solar wind, the light noble gases He and Ne of 566 the MMs from Dome C analyzed in this work are dominated by the trapped solar wind 567 component, while for heavy Kr and Xe a trapped "planetary" component (Ott, 2014) accounts 568 for most of the inventory. Argon generally represents a mixed case. Here we present a short 569 570 general overview of abundances, while in the following sections (4.2 and 4.3), we will discuss the individual cases in more detail. We show an overview of the trapped noble gas abundances 571 in Figure 5 (crystalline MMs, left; others, right). The concentrations of the major isotopes are 572 plotted, and, since MMs have often been compared to CM meteorites (Kurat et al., 1994; 573 Engrand and Maurette, 1998), we normalized them to the concentrations in CM meteorites, 574 where we considered CM Maribo #2 as reported in Haack et al. (2012) as representative, as we 575 576 did in Baecker et al. (2018). Again, the possible, but probably small, radiogenic contributions to ⁴He are ignored. Shown are the individual specimens as well as the geometric means for the 577 578 two populations. For comparison, we also show the geometric means for the TAM MMs of 579 Baecker et al. (2018) and for the Antarctic MMs collected at Dome Fuji and in the Yamato Mountain area as reported by Osawa et al. (2000), Osawa and Nagao (2002a, b) and Osawa et 580 al. (2003b). Data on another suite of Dome Fuji samples analyzed by Okazaki et al. (2015) are 581 shown separately, while, for the geometric mean of TAM MMs, we distinguish between 582 scoriaceous and unmelted MMs. 583



585 <u>Figure 5.</u> Noble gas concentrations in DC MMs, normalized to CM Maribo #2 (Haack et al., 2012). Shown are the individual specimens 586 as well as the geometric means for the two populations (Xtal and "other") separately. For comparison we also show the geometric means 587 for the TAM MMs of Baecker et al. (2018) and for Antarctic MMs collected at Dome Fuji and in the Yamato Mountain (Osawa et al., 588 2000; Osawa and Nagao, 2002a, b; Osawa et al., 2003b). Data the Dome Fuji samples of Okazaki et al. (2015) are shown separately, while 589 for the geometric mean of TAM MMs we distinguish between scoriaceous and unmelted MMs.

Several features are apparent. First, the scoriaceous MMs show, by far, the lowest noble 590 591 gas concentrations. This is to be expected given the higher temperature experienced by scoriaceous (partly melted) micrometeorites during atmospheric entry, resulting in more severe 592 gas loss. Nevertheless, concentrations also in the unmelted TAM MMs are lower than in the 593 other MMs, in particular for He and Ne. The larger sizes of the TAM MMs (~600 µm on 594 595 average; Baecker et al., 2018) compared to the Dome Fuji and Yamato Mountain MMs as well as our DC MMs, and the correspondingly lower surface to volume ratio of TAM MMs may 596 have played a role, since He and Ne are predominantly of solar wind origin and thus surface-597 correlated (implantation depth $< 0.1 \,\mu$ m). Second, in accordance with such an interpretation, 598 on average the concentrations of (volume-correlated) Xe in the unmelted TAM MMs are not 599 600 too different from those of the Dome Fuji and Yamato Mountains MMs of Osawa et al. (2000, 2003b) and Osawa and Nagao (2002a, b), as well as the reference CM meteorite, for that matter. 601 This similarity holds also in comparison with the majority of our DC MMs, although at least 602 two of the latter show significantly higher concentrations, raising the mean for our DC samples 603 above those reference abundances. 604

605 Third, the average concentrations reported by Okazaki et al. (2015) for MMs recovered from surface snow near Dome Fuji are one to two orders of magnitude higher than the others 606 reported previously, but are roughly matched by some of our DC MMs. The similarity between 607 the two sets of data is striking and most probably related to the collection method. Our Dome 608 C and the Dome Fuji particles of Okazaki et al. (2015) were collected using the same protocol, 609 which is gentle and rapid melting of surface snow (Duprat et al., 2007; Okazaki et al., 2015). 610 This protocol is specific to these 2 collections, where the particles were trapped in snow at 611 temperatures far below 0 °C before being extracted from their host matrix (the snow) rapidly 612 (within hours). In contrast, MMs from other collections were trapped in a matrix (ice, 613 614 sediments...) that experienced much larger temperature variations and mechanical stress. In particular, most MMs from the other collections experienced repeated contact with liquid water, 615 in particular during thawing/freezing cycles at the surface of ice fields, whereas the MMs from 616 Dome C and the Dome Fuji samples of Okazaki et al. (2015) experienced only limited contact 617 with liquid water, for a few hours during their extraction (note that this is also different for the 618 samples extracted from snow at Mount Fuji; Osawa et al., 2000; Osawa and Nagao, 2002a). 619 The striking difference in the noble gas concentrations between MMs obtained by different 620 collection methods suggests that part of the noble gases initially present in dust in interplanetary 621 space is hosted by a phase that may be subject to leaching by water. 622

Another interesting feature is the depletion of Xe relative to Kr and the reference CM meteorite ratio (roughly that in phase Q, the major carrier of heavy noble gases in primitive meteorites; Ott, 2014). We have argued previously that this may a sign for contamination by (terrestrial) air, which has an 84 Kr/ 132 Xe ratio of ~28, much higher than the primitive noble gas component in carbonaceous chondrites (~1). We will come back to this later (chapter 4.3).

628 4.2. Solar wind gases: Helium and Neon

The solar wind origin of the noble gases He and Ne is highlighted in Figure 6, which is a plot of trapped ⁴He vs. ²⁰Ne abundances for those AMMs, where both could be measured (Tables 2, 3 and 6). Minor corrections for cosmogenic contributions have been applied but are

- 632 negligible. Any radiogenic ⁴He present is still included, however. Compared to the ⁴He/²⁰Ne
- ratio of 656 determined for present-day solar wind (Heber et al., 2009), which defines the
- solid line in Figure 6, all data points are shifted down / to the right, i.e. have lower ${}^{4}\text{He}/{}^{20}\text{Ne}$.
- As noted before (chapter 4.1; and Baecker et al., 2018), unmelted MMs have higher
- 636 concentrations of solar wind gases than partially melted (scoriaceous) ones, and the
- 637 concentrations roughly scale with size, a consequence of the fact that they are surface-638 correlated. This is also apparent in that MM DC 06 09 11, which is partly scoriaceous
- (FgC/Sc), has lower concentrations and lower ⁴He/²⁰Ne than the "purely FgC" MMs.
- However, we do not observe this for the other FgC/Sc MM DC 06 09 189, which has the
- highest concentrations of 4 He and 20 Ne in our sample set.

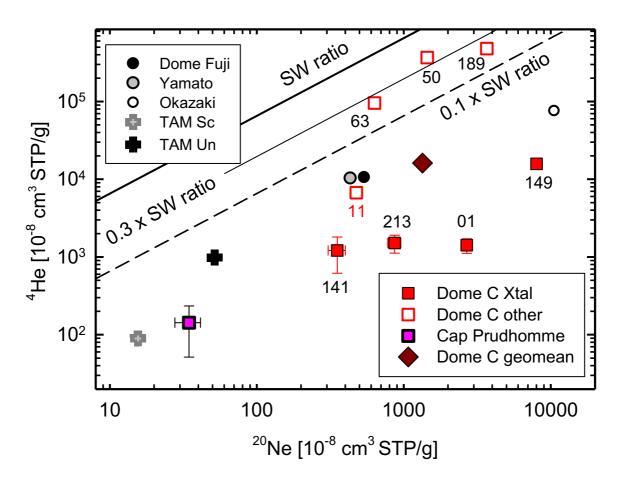


Figure 6: Concentrations of ⁴He plotted vs. concentrations of trapped ²⁰Ne. The individual 643 data points for our DC MMs are labeled with the last digits of the sample name (Table 1). We 644 also show the geometric means for our DC MMs as well as for the results of previous studies 645 of MMs from Dome Fuji, the Yamato Mountains area and the Transantarctic Mountains. 646 References: Osawa et al. (2000, 2003b), Osawa and Nagao (2002a, b), Baecker et al. (2018). 647 The Dome Fuji value from the Okazaki et al. (2015) study is shown separately, and the TAM 648 MMs are distinguished between Unmelted (Un) and scoriaceous (Sc). The lines correspond to 649 the solar wind ⁴He/²⁰Ne ratio (Heber et al., 2009), as well as 0.3x and 0.1x the solar wind 650 651 ratio.

- A new feature, not apparent from previous work, is the difference between MMs that 652 are crystalline (Xtal) and those that are fine-grained carbonaceous (FgC). While both cover 653 about the same range in concentrations of Ne, the Xtal ones have significantly lower ⁴He and 654 thus much lower ${}^{4}\text{He}/{}^{20}\text{Ne}$, by almost two orders of magnitude – geometric means are ~1.6 vs. 655 ~170, ignoring partly scoriaceous DC 06 09 11. Previous noble gas studies have mostly 656 distinguished between, on one hand, unmelted (often simply called MM; e.g., Osawa et al., 657 658 2003) and, on the other, scoriaceous MMs and completely melted cosmic spherules, thus missing the apparent difference between Xtal and FgC unmelted MMs. 659
- Compared to previous works, the mean ⁴He and ²⁰Ne concentrations for our Dome C 660 MMs as well as the MMs of Okazaki et al. (2015) are higher, but plot roughly along a line 661 parallel to the solar wind line, i.e. they all have, on average, similar ${}^{4}\text{He}/{}^{20}\text{Ne}$ ratios, of ${\sim}20$, 662 i.e. ~0.03 times the SW ratio. On the other hand, our samples clearly defined as FgC MMs, 663 show roughly ten times higher ratios of ~0.3x SW, as do some in the earlier studies (Osawa 664 and Nagao, 2002a; Osawa et al., 2000, 2003b; see also Fig. 2 in Okazaki et al., 2015). The 665 fact that, in all studies so far, the average ⁴He/²⁰Ne is about the same suggests that, in all these 666 studies, the mix of fine-grained carbonaceous and crystalline MMs was fairly similar. 667

Interestingly, ⁴He/²⁰Ne ratios observed in IDPs (Nier and Schlutter, 1990; Pepin et al., 668 2000) more or less cover the same range as those observed for unmelted MMs. We have 669 argued earlier (Baecker et al., 2018), based on comparison not only with IDPs, but also with 670 lunar soil samples and grains returned from asteroid Itokawa (Nagao et al., 2011), that ratios 671 such as those observed in MMs DC 06 09 50, 06 09 63 and 06 09 189 may be common 672 673 (characteristic) for the pre-terrestrial ratio in MMs. Even though lower than in solar wind proper, such ratios are therefore not necessarily indicative of loss during passage through the 674 terrestrial atmosphere. Differences in absolute concentrations (of both He and Ne) may then 675 be related to size (via the surface/volume ratio), fractional loss of the outermost ~0.05 nm, 676 677 where the solar wind gases reside, and - in cases where not the whole MM was used for noble 678 gas analysis - the unknown fraction of surface present in the sample actually used for the noble gas work (Baecker et al., 2018). 679

As for the difference in average ⁴He contents and ⁴He/²⁰Ne ratios between Xtal and FgC 680 MMs, it appears likely that this was established during atmospheric entry heating. The 681 apparent loss of He by one to two orders of magnitude suggested by Fig. 6 is similar to that 682 found by Füri et al. (2013) in their series 1 experiment for two-second heating to 1350 °C of 683 grains of the Orgueil (CI) meteorite, used for simulation of entry heating. After this, the grains 684 were still characterized as fine-grained, non-vesicular (Toppani et al., 2001). We note, 685 however, that in this case Füri et al. (2013) also observed significant loss of Ne (some 80 %), 686 while for our crystalline MMs there is no indication for Ne loss (Fig. 6). We also note that the 687 results of Füri et al. (2013) are compromised by large variations in the abundances of the 688 starting material used for heating and that there is no information on the abundance before 689 heating in the individual samples. In any case, the differences between the Xtal and other 690 grains of our study indicate that, besides the temperature reached during atmospheric entry, 691 692 the mineralogical makeup of the MM also determines the degree of loss of solar wind He. As a consequence, the temperature dependence of ⁴He release during analysis (Nier and 693

Schlutter, 1993; Okazaki et al., 2015) alone may not be a reliable measure for determining thedegree of heating experienced by a MM during atmospheric entry.

While in this work on MMs from Dome C we concentrated on unmelted MMs, it useful to keep in mind that even in scoriaceous MMs some solar wind Ne has been retained. Preferred orientation during atmospheric entry that resulted in a small fraction of surface to remain intact may be a possible explanation, but it cannot be excluded that some amount of solar wind gas might have been acquired by subunits of the MMs during residence in an asteroidal regolith prior to assembly and ejection. This could then reside in the MM's interior (Baecker et al., 2018) rather than close to the surface.

703 4.3. Heavy noble gases: Argon, krypton and xenon

The case of krypton and xenon is distinct from that of He and Ne since they can be expected to be primarily of the "planetary" type (Ott, 2014) and volume- rather than surfacesited. The case of Ar is more complicated and differs from that of our TAM samples (Baecker et al., 2018), since the DC MMs have significantly higher solar wind gases concentrations (chapter 4.2; Figs. 5 and 6).

The difference is also obvious in a plot of 36 Ar/ 132 Xe vs. 84 Kr/ 132 Xe (Fig. 7) where the high 36 Ar/ 132 Xe ratios in all MMs, with the exception 06_09_189, can only be explained by the contribution of solar components. As an example, treating the measured compositions as a 3-component mixture of solar wind, Q and air (all unfractionated), for DC 06_09_63 the contribution to 36 Ar of SW is 65% (vs. 24% from air and 11% from Q). For DC 06_09_11, as another example, the corresponding values are 57% (SW), 7% (air) and 35% (Q).

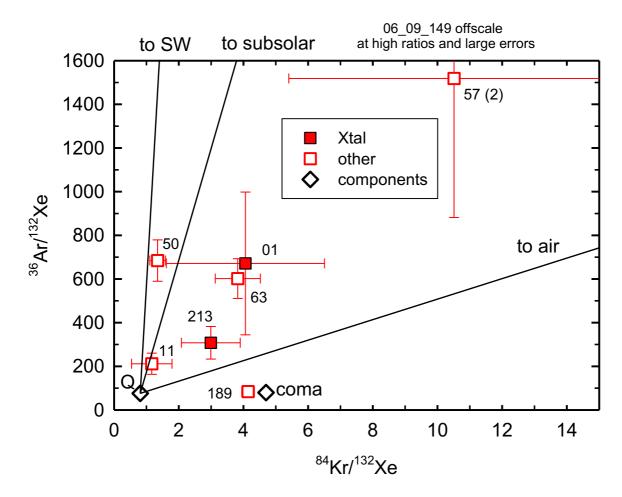
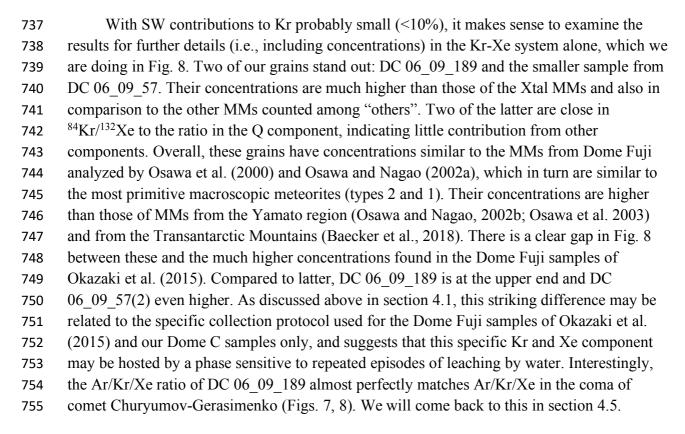
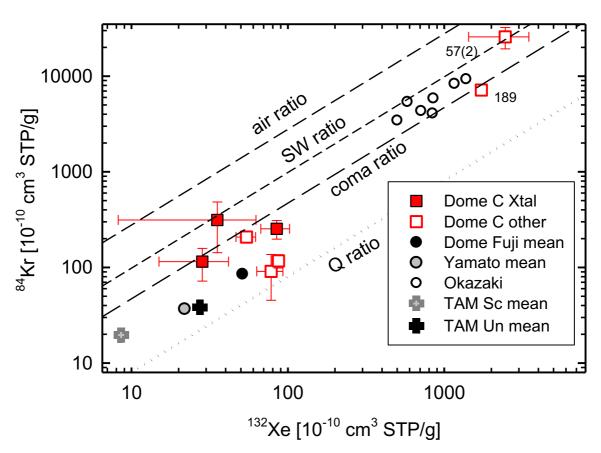


Figure 7. Plot of ³⁶Ar/¹³²Xe vs. ⁸⁴Kr/¹³²Xe. The data points are labeled with the last digits of
the sample names (Table 1); DC_06_09_149 plots off scale. The composition of Q
(Busemann et al., 2000; Ott, 2014) and of gases in the corona of comet ChuryumovGerasimenko (Rubin et al., 2018) are shown for reference, as well as mixing lines between Q
and solar wind (Vogel et al., 2011), subsolar gas (Ott, 2014) and air.

On the other hand, it is also obvious, in particular from DC 06 09 11, that a component 721 similar to Q, with low ratios, is needed as well. For Xe, in the three-component mixing model 722 discussed above. O is always the dominant component, accounting for 88 and 98% in the two 723 examples. As a result, the Xe isotopic compositions discussed in chapters 3.3 and below in 724 725 chapter 4.5, are essentially those of the non-solar component(s). For Kr, the situation is often 726 not so clear-cut, but from the 3-component exercise it still follows that the SW contribution is comparably small (usually <10%). Of course, the simple 3-component assumption used in the 727 estimates above is somewhat naive, with some of the components being possibly elementally 728 fractionated compared to their normal composition, and with gases that have Ar/Kr/Xe ratios 729 like in the coma of comet Churyumov-Gerasimenko (Rubin et al., 2018) as a possible 730 alternative (to air) or as an additional component. Still, the exercise provides some clues. 731 Interestingly, but possibly just by coincidence, the position of DC 09 06 189 is identical to 732 that of the coma composition. This special MM, which is fine-grained carbonaceous (FgC), 733 but also partially scoriaceous, does not require a contribution from solar wind Ar (nor Kr or 734 Xe, for that matter), which contrasts with the fact that it has the highest abundances of (solar) 735 He and Ne (Fig. 6) and a 20 Ne/ 36 Ar ratio very much like the others (Table 3). 736





⁷⁵⁷ <u>Figure 8.</u> Plot of ⁸⁴Kr vs. ¹³²Xe concentrations. DC 06_09_189 and DC 06_09_57(2) with the

highest abundances are labeled. For comparison geometric means from previous studies on
MMs from Mount Fuji, the Yamato Mountains and the Transantarctic Mountains (TAM) are
shown (Osawa et al. ,2000, 2003b; Osawa and Nagao, 2002a, b; Baecker et al. 2018). The
Dome Fuji MMs of Okazaki et al. (2015) are shown as individual data points and are not
included in the Dome Fuji mean, while the TAM MMs are distinguished between Unmelted
(Un) and scoriaceous (Sc). The various lines correspond to the ⁸⁴Kr/¹³²Xe ratio in air, SW, the

coma of comet Churyumov-Gerasimenko and Q. References as in Figure 7.

766 4.4. Cosmic ray exposure (CRE) and provenance

Cosmic ray exposure ages are commonly calculated from the overabundance of isotopes 767 that are much rarer in trapped components than the other isotopes of the same element, but are 768 produced by cosmic ray interaction in roughly equal amounts, such as ³He, ²¹Ne and ³⁸Ar. 769 Their concentrations – calculated as described in chapter 3.4 - are listed in Table 6. The 770 favored approach is usually that using ²¹Ne, because this isotope is less susceptible to loss 771 than ³He and less sensitive to variations in chemical composition than ³⁸Ar. The problem in 772 using ²¹Ne with MMs (as well as IDPs, for that matter) is the high abundance of solar wind 773 Ne (and solar wind He, for the case of ³He). Shifts to the right in the Ne three-isotope plot of 774 Fig. 2 indicative of cosmogenic ²¹Ne are therefore small and often zero within 2 σ . This is 775 different from the situation in our TAM MMs (Fig. 2), which have generally much lower 776 concentrations of solar wind noble gases (Baecker et al., 2018). It is similar, however, to the 777 situation for MMs from the Yamato Mountains and Dome Fuji (Osawa and Nagao, 2002a, 778 2002b; Osawa et al., 2000, 2003): no cosmogenic Ne was detected in the Yamato Mountains 779 MMs, and only 10% of the Dome Fuji MMs showed detectable ²¹Ne excesses (Osawa et al., 780 2003), mostly such with high 20 Ne/⁴He ratios (see their Fig. 5). 781

From the nominal amounts of cosmogenic ²¹Ne (Table 6; chapter 3.4), we have 782 calculated nominal cosmic ray exposure ages and inferred places of origin by considering the 783 Poynting-Robertson effect of small particles spiraling towards the Sun (Wyatt and Whipple, 784 785 1950; Trappitsch and Leya, 2013). The calculations, the results of which are listed in Table 7, were obtained using the Trappisch and Leya (2013) cosmic ray production rates, with updates. 786 Two values are listed in each case, corresponding to the two approaches for defining the 787 trapped composition used in Table 6 and described in chapter 3.4. We have done the same 788 using ³He (also two sets of values corresponding to the two values in Table 6). For this, we 789 corrected ³He assuming recoil losses of 32% (including losses of directly produced ³He and 790 the larger losses of ³H produced as a precursor), which seems roughly appropriate for particles 791 in the studied size range (Ott et al., 2009; Trappitsch and Leya, 2016). No values are given for 792 the cases (²¹Ne in DC 06 07 213; ³He in DC 06 09 50 and DC 06 09 149) that result in 793 negative nominal values (see also chapter 4.5). Note also that, apart from the analytical and 794 production rate uncertainties, the action of the Poynting-Robertson effect may be more 795 complicated than in this simple approach, in particular in case of non-spherical and / or fluffy 796 particles (Kimura et al., 2002). Moreover, the approach considers only the Poynting-797 Robertson effect and not others that may affect the orbit of a particle. 798

The most convincing case is that of DC 06_08_01 , which results in a CRE age of ~2 Ma and a place of origin of ~4 au, i.e. well in the asteroid belt. Notably, ³He gives consistent results, indicating that any loss of cosmogenic ³He has been small. Interestingly, this is an

Xtal MM characterized by low concentration of solar wind He, relative to solar wind Ne 802 (chapter 4.2; Fig. 5). On the one hand, the relatively low background of solar wind He leads to 803 a relatively high ${}^{3}\text{He}/{}^{4}\text{He}$ ratio of ${}^{-14}$ x 10⁻⁴ (Table 2) and thus allows identifying rather 804 clearly the abundance of cosmogenic ³He (Fig. 2). On the other hand, low ⁴He with high ³He 805 also demonstrates the different loss behavior of surface-sited solar wind vs. cosmogenic He 806 contained in the bulk of the MM. Consistent results from ²¹Ne and ³He are also obtained for 807 808 Fg/Sc DC 06 09 11, however with a very short inferred exposure age of only ~0.2 Ma with an origin at ~1.5 au. Ages from ²¹Ne are also short for DC 06 09 50 and DC 06 09 63, 809 while DC 06 09 189 (Fg/Sc) is similar to DC 06 08 01, i.e. compatible with an origin in the 810 asteroid belt. The same is true for the other case of "safe" cosmogenic ³He, DC 06 07 213 811 (Fig. 1), with an inferred CRE age of \sim 1 Ma, and coming from \sim 3 au. No ²¹Ne age could be 812 obtained for this MM, however. Finally, a rather long cosmic ray exposure from ²¹Ne is 813 indicated for DC 06 09 149, which has the smallest analytical uncertainty and is clearly 814 offset from the trapped composition lines in Fig. 2. The nominal age of ~15 Ma, still with 815 816 comparably large uncertainty, suggests an origin at a much larger distance, possibly beyond Saturn, thus from a cometary source. This age (like all the other nominal ages) rests on the 817 assumption that the cosmic ray exposure took place during travel as a small particle. 818 However, it cannot be ruled out that much (or even most) cosmogenic ²¹Ne production took 819 place in the parent body regolith instead (Trappitsch and Leya, 2013). For GCR production in 820 a well-mixed regolith the ²¹Ne production rate has been estimated as 0.11x10⁻⁸ cm³ STP per 821 gram and Ma Trappitsch and Leva, 2013), which in case of DC 06 09 149 would mean an 822 exposure roughly 100 to 150 Ma. 823

Table 7. Nominal cosmic ray exposure (CRE) ages and implied places of origin (distance 825 from the sun) of Dome C MMs. These have been calculated following Trappitsch and Leva 826 (2013) from the interplay between cosmic ray production and action of the Poynting-827 Robertson effect. The approach assumes ²¹Ne and ³He excesses over abundances in common 828 trapped components are due to cosmic ray production, as described in the text and listed in 829 Table 6, with an assumed 32% recoil loss of cosmogenic ³He (see text). For production by 830 solar cosmic rays updated production rates for a rigidity of 100 MV have been used. Chemical 831 abundances used for the calculation are CM chondritic. Column 2 gives the type of MM 832 (Table 1). In the calculations masses for the complete MMs three times those of the analyzed 833 specimens were assumed, with correspondingly larger sizes. Values in italics / small print are 834 suspicious and the observed effects may require an explanation other than cosmic ray 835

sample	type	T ₂₁ (Ma)	r ₂₁ (au)	T ₃ (Ma)	r ₃ (au)
DC 06_07_213	Xtal			1.1 (-0.3, +0.3) 0.9 (-0.3, +0.3)	3.3 (-0.4, +0.4) 3.0 (-0.5, +0.4)
DC 06_08_01	Xtal	2.52 (-1.42, +1.94) 1.50 (-0.89, +1.31)	4.64 (-1.50, +1.46) 3.63 (-1.18, +1.26)	2.1 (-0.3, +0.3) 2.0 (-0.3, +0.3)	4.3 (-0.3, +0.3) 4.1 (-0.3, +0.3)
DC 06_09_11	FgC/Sc	0.20 (-0.13, +0.19) 0.15 (-0.11, +0.18)	1.62 (-0.37, +0.44) 1.50 (-0.34, +0.43)	0.3 (-0.3, +0.7) 0.1 (-0.1, +0.6)	1.9 (-0.9, +1.1) 1.4 (-0.4, +1.2)
DC 06_09_50	FgC	0.11 (-0.06, +0.06) 0.07 (-0.04, +0.04)	1.28 (-0.14, +0.14) 1.17 (-0.09, +0.11)		
DC 06_09_63	FgC/Xtal	0.05 (-0.04, +0.06) 0.03 (-0.03, +0.05)	1.14 (-0.12, +0.14) 1.08 (-0.08, +0.13)	16.8 (-5.9, +6.0) 14.7 (-4.0, +4.0)	10.2 (-2.0, +1.7) 9.5 (-1.4, +1.2)
DC_06_09_149	Xtal	17.2 (-10.3, +10.8) 12.3 (-8.9, +9.9)	14.6 (-5.3, +4.0) 12.4 (-5.8, +4.2)		
DC 06_09_189	FgC/Sc	2.41 (-1.62, +2.35) 1.30 (-0.95, +1.62)	4.78 (-1.93, +1.87) 3.57 (-1.54, +1.67)	26.1 (-9.2, +9.3) 16.6 (-14.2, +14.2)	15.5 (-3.0, +2.4) 12.3 (-7.5, +4.5)

exposure, as discussed in the text (chapter 4.4).

837

There are several additional results that are difficult to understand (or to accept, for that 838 matter). As for ³He ages, these concern DC 06 09 63 and DC 06 09 189. The latter can be 839 easily explained, though, and will disappear if we allow for a slightly higher ³He/⁴He ratio for 840 the trapped component (>3.26 x 10^{-4} instead of (2.85 ± 0.12) x 10^{-4} and (3.00 ± 0.20) x 10^{-4} in 841 the two approaches will do). DC 06 09 63 is not as easily explained away. Although the 842 measured ${}^{3}\text{He}/{}^{4}\text{He}$ of (4.14 ± 0.30) x 10⁻⁴ is lower than the ratio in the SW proper, it is still 843 much higher than in the FSW component and the mixing approach (alternative 1) suggests a 844 trapped ratio of $(2.83 \pm 0.40) \times 10^{-4}$. It is also much higher than in the MMs with the highest 845 SW concentration, where the influence of the cosmogenic component must be suppressed. 846 The lack of equivalent amounts of cosmogenic Ne suggests a non-cosmogenic origin for the 847 ³He (if real), and similarly excesses of ³He not easily attributed to cosmic ray production have 848 been found in some IDPs (Nier and Schlutter, 1993; Pepin et al., 2000, 2001). The more 849 distinct excesses in these studies, with measured ³He/⁴He often clearly above the SW value, 850 would require unreasonably long CRE times of between 50 Ma and >10 Ga, and are, as in our 851

case, characterized by lack of corresponding excesses of cosmogenic ²¹Ne. At this time, we
cannot add further to the extensive discussions of possible alternative explanations by Pepin et
al. (2000, 2001), but none of them seems totally convincing.

855 4.5. Cometary origins (?)

856 The smaller IDPs collected in the stratosphere most probably have a cometary origin, in particular for the chondritic porous (CP) subtype (e.g., Bradley, 2005; Ishii et al., 2008; 857 Messenger et al., 2003; Busemann et al., 2009). In contrast, micrometeorites recovered from 858 ice and snow, from traps, or fossil have generally been regarded as of asteroidal origin and as 859 largely related to common meteorites (Kurat et al., 1994; Genge et al., 1997, 2020; Taylor et 860 al., 2012; Cordier and Folco, 2014; Noguchi et al., 2015; Shyam Prasad et al., 2018), 861 including differentiated ones (Taylor et al., 2007; Cordier et al., 2012). The large deuterium 862 excesses (Duprat et al., 2010) and the high nitrogen content of their organic matter (Dartois et 863 864 al., 2013; Yabuta et al., 2017) is suggestive of a cometary origin for the ultra-carbonaceous MMs, but otherwise the question whether at least some MMs are cometary has loomed large 865 from the start of MM studies (e.g., Noguchi et al., 2015). Still, the modelling by Nesvorný et 866 al. (2010), see also Carrillo-Sánchez et al. (2020) and (Rojas et al, 2021), suggests that a 867 significant part of the cosmic dust reaching Earth in the size range of the MMs may derive 868 from Jupiter Family Comets (JFC). 869

870 Noble gases can contribute in this matter, in different ways. Particles from asteroidal and cometary sources may have, on average, different entry velocities when encountering 871 Earth, and, in the study of IDPs, Nier and Schlutter (1990, 1993) tried to use the release 872 profile of solar wind He as a measure. In principle, a similar approach might be applicable to 873 MMs, however, as we have shown in chapter 4.1, also the mineralogical makeup plays a role 874 for the observed He abundance: Xtal MMs in our collection have much lower ⁴He abundances 875 and ⁴He/²⁰Ne ratios than FgC MMs. While we have not studied the release pattern in this 876 work, presumably it is also different between the two types, which, if not taken into account, 877 tends to obscure the entry effect. Still, it may be a useful approach in future work. 878

Another approach can be based on differences in cosmic ray exposure. In our case, and 879 880 presumably in all cases involving primitive small particles, this is analytically challenging because of the large abundances of solar wind gas, with only minor and barely (if at all) 881 detectable excesses of cosmogenic ³He and ²¹Ne, with the added problem that cosmogenic 882 gases (like the trapped gases) may have been lost during atmospheric entry (e.g., Füri et al., 883 2013; Baecker et al., 2018). In our data set, nominal CRE ages that we were able to obtain are 884 all consistent with an asteroidal source, with the possible exception of DC 06 09 149 and the 885 exception of the possible excess ³He in DC 06 09 63 (chapter 4.4). They do not, however, 886 rule out a cometary origin, as once ejected from their parent body, cometary dust particles are 887 subject to non-gravitational forces that affect their initial velocities and trajectories and may 888 889 result in orbits close to those expected from asteroidal dust.

A third approach can be based on the search for signatures in the trapped noble gas
components that have been found in *bona fide* cometary materials. There are two sets of data
that we can relate to in this approach, which have sampled different comets (though both

Jupiter Family) and different cometary reservoirs: the mass spectrometer onboard the Rosetta
mission measured the composition of noble gases in the coma of comet 67P/ChuryumovGerasimenko (Marty et al., 2017; Rubin et al., 2018), while He and Ne abundances were
measured in the laboratory in solids captured from the coma of 81P/Wild 2 in aerogel and
returned to Earth (Marty et al., 2008). The former presumably measured gases that were
trapped in ices (maybe primarily CO₂ ice; Rubin et al., 2018), while the latter analysis was of
solid particles, probably high-temperature igneous grains (Marty et al., 2008).

In fact, we do find some similarities between the observed signatures, which, however, 900 are suggestive only. Two of our MMs show similarities to features observed in the coma of 901 67P/Churyumov-Gerasimenko. For one, the elemental abundance ratios ³⁶Ar/¹³²Xe and 902 ⁸⁴Kr/¹³²Xe for DC 06 09 189 (Figs. 7 and 8) are almost identical to what has been inferred 903 for the bulk abundance ratios of the coma gases (Rubin et al., 2018). Given that a substantial 904 905 fraction of ³⁶Ar is probably from solar wind, this must appear to be fortuitous, at least as far as ³⁶Ar/¹³²Xe is concerned. One might also expect some elemental fractionation between noble 906 gases trapped in ice vs. other, more refractory, solids. 907

More telling than elemental ratios are isotopic ratios, and the second similarity is in the 908 isotopic composition of Xe and of Ar. Xe as measured in the coma of Churyumov-909 910 Gerasimenko (Marty et al., 2017) is highly diagnostic, i.e. characterized by huge depletions of the two heaviest isotopes, ¹³⁴Xe and ¹³⁶Xe, when compared to other Solar System reservoirs. 911 A hint for a contribution from such a component is seen in the Xe pattern of the small MM 912 fraction DC 06 09 57(2) shown in Fig. 4, and in a more extended version in Fig. 9. But this 913 case has been analytically challenging, with an amount of 132 Xe of only $\sim 2x10^{-15}$ cm³ STP 914 (some 50,000 atoms) after blank correction (¹³⁶Xe: ~8600 atoms), and where the blank 915 contributed about half of the measured ¹³²Xe (2/3 in case of ¹³⁶Xe). To illustrate the point, we 916 show in Fig. 9 also the ratios as measured, before blank subtraction. In essence, while 917 suggestive, the measurement errors, both from the MM and Rosetta, are too large for a firm 918 919 conclusion. It is intriguing, though, that in the same MM sample we also measured highly unusual Ar, with ${}^{38}\text{Ar}/{}^{36}\text{Ar} = 0.1495 \pm 0.039$, significantly lower than in any other well-920 defined trapped Ar component (chapter 3.2). The corresponding ${}^{36}\text{Ar}/{}^{38}\text{Ar}$ ratio of 6.69 ± 0.17 921 agrees within uncertainties with the coma ${}^{36}\text{Ar}/{}^{38}\text{Ar}$ ratio of 5.4 ± 1.4 reported by Rubin et al. 922 923 (2018), however, in this case, the large error of the comet measurement prevents any firm conclusion. In addition, the Ar/Xe and Kr/Xe ratios for DC 06 09 57(2) are significantly 924 larger than reported for coma gas (Fig. 7), so strong elemental fractionation would be 925 926 required.

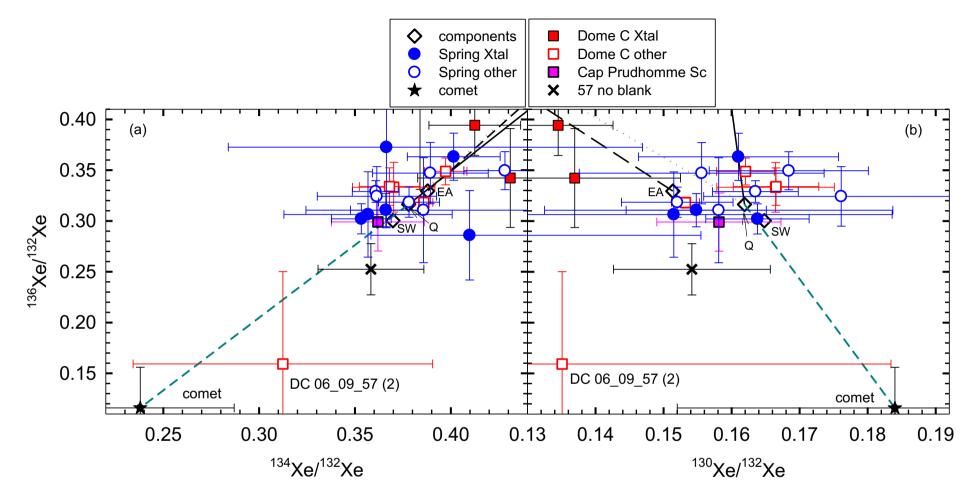


Figure 9. Same as Figure 4, but with scale expanded to include DC $06_{09}_{57(2)}$ and the composition measured in the coma of comet Churyumov-Gerasimenko (Marty et al., 2017). Also shown is the measured value for DC 06 09 57(2), without extraction blank subtracted ("57 no blank").

The signatures discussed above are likely those of the "icy" cometary component, but, 931 932 in the MMs, we are dealing with more refractory solids. So, if a connection to our MMs actually exists, there must have been some equilibration between noble gases in cometary ice 933 and cometary solids. Less surprising might be to find, in grains of cometary origin, the 934 signature that has been observed in grains returned by the Stardust mission. The diagnostic 935 936 feature here is the isotopic composition of Ne, which resembles Ne in the Q phase of chondritic meteorites (Marty et al., 2008). In fact, the same signature appears to have been 937 seen recently in a refractory IDP, Manchanito (Ogliore et al., 2020), for which also other 938 properties point toward a cometary origin. Data for this IDP, as well as the Stardust grain 939 data, are plotted together with our MM data in the Ne three-isotope plot of Fig. 2b. Obviously, 940 two of our Xtal grains, DC 06 07 213 and DC 06 09 141 (both with relatively low ²⁰Ne 941 abundance), plot at a similar position, albeit at slightly higher ²⁰Ne/²²Ne, probably due to 942 contribution from SW Ne. Presence of Q as a trapped noble gas component is certainly 943 consistent with the Xe isotopic composition we observe, not only in these, but also most other 944 MMs (Fig. 4; chapter 3.3). but the ²⁰Ne/¹³²Xe ratio for these MMs is on the order of 1000 or 945 higher, i.e. much higher than the ratio of 3 found for Q gas in carbonaceous chondrites 946 (Busemann et al., 2000; Ott, 2014). Thus it is unlikely to be hosted by the Q phase as known 947 from macroscopic meteorites that come from asteroids. It is also opposite the direction 948 949 expected in case of partial loss of Q-phase noble gases. Obviously, although the analytical errors do not allow to firmly equate the observed composition with that of Q-Ne, the same 950 holds for the Manchanito particle and Stardust grain data. Having data for Xe (or the complete 951 noble gas spectrum) in Manchanito and Stardust grains would have been useful in this 952 context. Still, the similarity between the data is intriguing, and with the Stardust grains 953 954 undoubtedly cometary, the two MMs and the Manchanito IDP may very well carry cometary signatures. 955

956

957 5. Summary - Conclusions

We have performed a complete noble gas study on fragments from ten Antarctic 958 micrometeorites (AMMs) collected at Dome C, including the isotopes of Kr and Xe, which 959 have been largely missing in previous noble gas studies of micrometeorites. Additional data 960 obtained on a scoriaceous MM from Cap Prudhomme do not provide much information. In 961 the Dome C MMs, He and Ne are dominated by a solar component, with lower ⁴He 962 concentrations and ⁴He/²⁰Ne ratios in crystalline (Xtal) compared to fine-grained 963 carbonaceous (FgC) MMs. The latter fall in the high range of solar wind noble gas 964 concentrations seen in earlier work, which may be related to the collection method (gentle and 965 rapid melting of surface snow) and indicate existence of a noble gas host phase in MMs that 966 967 may be affected by exposure to liquid water. The abundances of (volume-correlated) heavier gases are similar to what was found in previous studies of MMs. For this reason, solar wind is 968 also an important part of Ar and may make also a noticeable contribution to Kr in some cases, 969 however not to Xe. 970

971 Xe isotopic ratios are generally in between the composition dominating in primitive
972 macroscopic meteorites (Q component) and air, but, in most cases, uncertainties are too large

for strong conclusions. Because of the high abundances of SW He and Ne, cosmogenic
contributions are difficult to resolve. Nevertheless, when resolvable, cosmic ray exposure
ages based on cosmogenic ²¹Ne in combination with the Poynting-Robertson effect are
consistent with an origin from the asteroid belt in most cases. The same is true for the rarer
cases where we could also derive CRE ages from ³He. One of the MMs (of FgC/Xtal variety)
shows a ³He excess similar to what has been seen in some cluster IDPs.

979 There are hints that some of our (non-representative sample of DC) MMs may relate to 980 a cometary source, but uncertainties are too large for a firm conclusion. These include

- A Xtal MM, for which the abundance of cosmogenic ²¹Ne suggests it may 981 originate from beyond the orbit of Saturn. 982 A small specimen taken from a FgC MM that may contain Xe deficient in the 983 heavy isotopes ¹³⁴Xe and ¹³⁶Xe, like in the Xe component identified in the coma 984 of comet 67P/Churyumov-Gerasimenko, where it is probably derived from 985 cometary ice. The same MM has a precisely measured highly unusual ³⁶Ar/³⁸Ar 986 ratio of ~6.7, consistent with the measurement of coma Ar (${}^{36}\text{Ar}/{}^{38}\text{Ar} = 5.4 \pm$ 987 1.4). In this case, the large error of the coma measurement is an obstacle for a 988 firm conclusion. 989 Two MMs of Xtal type, with low SW contributions, that show Ne similar to 990 what has been found in laboratory analysis of refractory grains captured from 991 the coma of comet 81P/Wild 2 by the Stardust mission, and also more recently 992 in the IDP Manchanito, where this was considered as suggesting a cometary 993 origin., While trapped Ne with such a composition is also characteristic of the Q 994 component found in macroscopic meteorites, a connection to Q-Ne (and thus a 995 chondritic parent body) is unlikely because the Ne/Xe ratio (not obtained in the 996 Stardust and Manchanito analyses) differs by orders of magnitude. 997
- 998
- 999 Declaration of Competing Interest
- 1000

The authors declare no competing interests.

1001

1002 Acknowledgments

Siegfried Herrmann and Reinhard Haubold provided technical assistance with noble gas 1003 mass spectrometry. B.B., U.O. and M.T. acknowledge funding by the German Research 1004 Foundation, DFG-grants OT 171/5-1 and 5-2, M.T. also thanks the Klaus Tschira Stiftung. 1005 J.D. and C.E. were financially supported by ANR (Project COMETOR 18-CE31-0011), 1006 1007 Region Ile de France (DIM-ACAV+), PNP/INSU, CNES, IN2P3 and Labex P2IO. Collection of the micrometeorites was performed at CONCORDIA Station (project #1120) and 1008 financially supported by the French Polar Institute (IPEV). We are grateful to the staff of the 1009 French (IPEV) and Italian (PNRA) Polar Institutes for logistical help in the field. 1010

References:

1014 1015 1016	 Albarede F., Ballhaus C., Blichert-Toft J., Lee CT., Marty B., Moynier F. and Yin QZ. (2013) Asteroidal impacts and the origin of terrestrial and lunar volatiles. <i>Icarus</i> 222, 44-52. <u>https://doi.org/10.1016/j.icarus.2012.10.026</u>.
1017	Alexander C. M. O'D., Bowden R., Fogel M. L., Howard K. T., Herd C. D. K. and Nittler L.
1018	R. (2012) The provenances of asteroids, and their contributions to the volatile
1019	inventories of the terrestrial planets. <i>Science</i> 337, 721-723.
1020	https://doi.org/10.1126/science.1223474.
1021	Baecker B. (2014) Primordial and other noble gases in micrometeorites. PhD Thesis,
1022	University of Heidelberg, Germany, 362 p.
1023 1024 1025	 Baecker B., Ott U., Cordier C., Folco L., Trieloff M., van Ginneken M. and Rochette P. (2018) Noble gases in micrometeorites from the Transantarctic Mountains. <i>Geochim.</i> <i>Cosmochim. Acta</i> 242, 266-297. <u>https://doi.org/10.1016/j.gca.2018.08.02</u>.
1026	Basford J. R., Dragon J. C., Pepin R. O., Coscio Jr. M. R. and Murthy V. R. (1973) Krypton
1027	and xenon in lunar fines. <i>Proc. 4th Lunar Sci. Conf. (Suppl. 4, Geochim. Cosmochim</i>
1028	<i>Acta</i>), 1915-1955.
1029	Bekaert D. V., Avice G., Marty B., Henderson B. and Gudipati, M. S. (2017) Stepwise
1030	heating of lunar anorthosites 60025, 60215, 65315 possibly reveals an indigenous noble
1031	gas component on the Moon. <i>Geochim. Cosmochim. Acta</i> 218, 114-131.
1032	https://doi.org/10.1016/j.gca.2017.08.041.
1033 1034 1035	Benkert JP., Baur H., Signer P. and Wieler R. (1993) He, Ne, and Ar from the solar wind and solar energetic particles in lunar ilmenites and pyroxenes. <i>J. Geophys. Res.</i> 98 , 13,147-13,162. <u>https://doi.org/10.1029/93JE01460</u> .
1036	Bradley J .P. (2005) Interplanetary dust particles. In: Davis, A.M. (Ed.), Treatise in Geo-
1037	chemistry, vol.1. Elsevier–Pergamon, Amsterdam, pp.689-711.
1038	Bradley J. P., Ishii H. A., Gillis-Davis J. J., Ciston J., Nielsen M. H., Bechtel H. A. and
1039	Martin M. C. (2014) Detection of solar-wind produced water in irradiated rims on
1040	silicate minerals. <i>Proc. Natl. Acad. Sci.</i> 111 , 1732-1735.
1041	<u>https://doi.org/10.1073/pnas.132011511</u> .
1042 1043 1044	Britt D. and Consolmagno G. (2003) Stony meteorite porosities and densities: A review of the data through 2001. <i>Meteoritics & Planetary Science</i> 38 , 1161-1180. <u>https://doi.org/10.1111/j.1945-5100.2003.tb00305.x</u> .
1045	Busemann H., Baur H. and Wieler R. (2000) Primordial noble gases in "phase Q" in
1046	carbonaceous and ordinary chondrites studied by closed system stepped etching.
1047	<i>Meteorit. Planet. Sci.</i> 35, 949-973. <u>https://doi.org/10.1111/j.1945-5100.2000.tb01485.x</u> .
1048 1049	Busemann H., Nguyen A. N., Cody G. D., Hoppe P., Kilcoyne A. L. D., Stroud R. M., Zega T. J. and Nittler L. R. (2009) Ultra-primitive interplanetary dust particles from the

- comet 26P/Grigg–Skjellerup dust stream collection. *Earth Planet. Sci. Lett.* 288, 44–57.
 https://doi.org/10.1016/j.epsl.2009.09.007.
- 1052 Carillo-Sánchez J. D., Gómez-Martin H. C., Bones D. L., Nesvorný D., Pokorný P., Benna
 1053 M., Flynn G. J. and Plane J.M.C. (2020) Cosmic dust fluxes in the atmospheres of
 1054 Earth, Mars, and Venus. *Icarus* 335, #113395.
 1055 https://doi.org/10.1016/j.icarus.2019.113395.
- Cartwright J. A., Ott U., Herrmann S. and Agee C. B. (2014) Modern atmospheric signatures
 in 4.4 Ga Martian meteorite NWA 7034. *Earth Planet. Sci. Lett.* 400, 77-87.
 https://doi.org/10.1016/j.epsl.2014.05.008.
- 1059 Cordier C. and Folco L. (2014) Oxygen isotopes in cosmic spherules and the composition of
 1060 the near Earth interplanetary dust complex. *Geochim. Cosmochim. Acta* 146, 18-26.
 1061 <u>https://doi.org/10.1016/j.gca.2014.09.038</u>.
- Cordier C., Folco L., Suavet C., Sonzogni C. and Rochette P. (2011a) Major, trace element and oxygen isotope study of glass cosmic spherules of chondritic composition: The record of their source material and atmospheric entry heating. *Geochim. Cosmochim. Acta* 75, 5203-5218. <u>https://doi.org/10.1016/j.gca.2011.06.014</u>.
- Cordier C., Folco L. and Taylor S. (2011b) Vestoid cosmic spherules from the South Pole
 Water Well and Transantarctic Mountains (Antarctica): A major and trace element
 study. *Geochim. Cosmochim. Acta* 75, 1199-1215.
 https://doi.org/10.1016/j.gca.2010.11.024.
- Cordier C., Suavet C., Folco L., Rochette P. and Sonzogni C. (2012) HED-like cosmic
 spherules from the Transantarctic Mountains, Antarctica: Major and trace element
 abundances and oxygen isotopic compositions. *Geochim. Cosmochim. Acta* 77, 515 <u>529. https://doi.org/10.1016/j.gca.2011.10.021</u>.
- 1074 Cordier C., Baecker B., Ott U., Folco L. and Trieloff M. (2018) A new type of oxidized and
 1075 pre-irradiated micrometeorite. *Geochim. Cosmochim. Acta* 233, 135-158.
 1076 <u>https://doi.org/10.1016/j.gca.2018.04.010</u>.
- 1077 Cremonese G., Borin P., Martellato E., Marzari F. and Bruno M. (2012) New calibration of
 1078 the micrometeoroid flux on Earth. *Astrophys. J. Lett.* 749, L40.
 1079 https://doi.org/10.1088/2041-8205/749/2/L40.
- 1080 Crowther S., Mohapatra R. K., Turner G., Blagburn D., Kehm K., and Gilmour J. D. (2008)
 1081 Characteristics and applications of RELAX, an ultrasensitive resonance ionization mass
 1082 spectrometer for xenon. J. Anal. At. Spectrom. 23, 938-947.
 1083 <u>https://doi.10.1039/B802899K</u>.
- Dartois E., Engrand C., Brunetto R., Duprat J., Pino T., Quirico E., Remusat L., Bardin N.,
 Briani G., Mostefaoui S., Morinaud G., Crane B., Szwec N., Delauche L., Jamme F.,
 Sandt Ch. and Dumas P. (2013) UltraCarbonaceous Antarctic micrometeorites,
 probing the Solar System beyond the nitrogen snow-line. *Icarus* 224, 243-252.
 https://doi.org/10.1016/j.icarus.2013.03.002.

- Dartois E., Engrand C., Duprat J., Godard M., Charon E., Delauche L., Sandt C. and
 Borondics F. (2018) Dome C ultracarbonaceous Antarctic micrometeorites. Infrared and
 Raman fingerprints. *Astron. Astrophys.* 609, A65. <u>https://doi:10.1051/0004-</u>
 6361/201731322.
- Duprat J., Engrand C., Maurette M., Kurat G., Gounelle M. and Hammer C. (2007)
 Micrometeorites from central Antarctic snow: The CONCORDIA collection. *Adv.Space Res.* 39, 605-611. <u>https://doi.org/10.1016/j.asr.2006.05.029</u>.
- Duprat J., Dobricâ E., Engrand C., Aléon J., Marrocchi Y., Mostefaoui S., Meibom A.,
 Leroux H., Rouzaud J.-N. and Gounelle M. (2010) Extreme Deuterium excesses in
 ultracarbonaceous micrometeorites from Central Antarctic snow. *Science* 328, 742 745. <u>https://doi.org/10.1126/science.1184832</u>.
- Eberhardt P., Eugster O., and Marti K. (1965) A redetermination of the isotopic composition
 of atmospheric neon. Z. Naturforsch. 20a, 623-624. <u>https://doi.org/10.1515/zna-1965-</u>
 0420.
- Engrand C. and Maurette M. (1998) Carbonaceous micrometeorites from Antarctica.
 Meteorit. Planet. Sci. 33, 565-580. <u>https://doi.org/10.1111/j.1945-5100.1998.tb01665.x.</u>
- Flynn G. J. (1996) The delivery of organic matter from asteroids and comets to the early
 surface of Mars. *Earth, Moon, and Planets* 72, 469-474.
 https://doi.org/10.1007/BF00117551.
- Folco L., Rochette P., Perchiazzi N., D'Orazio M., Laurenzi M. A. and Tiepolo M. (2008)
 Microtektites from Victoria Land Transantarctic Mountains. *Geology* 36, 291-294.
 <u>https://doi.org/10.1130/G24528A.1</u>.
- Füri E., Aléon-Toppani A., Marty B., Libourel G. and Zimmermann L. (2013) Effects of atmospheric entry heating on the noble gas and nitrogen content of micrometeorites. *Earth Planet. Sci. Lett.* 377-378, 1-12. <u>https://doi.org/10.1016/j.epsl.2013.07.031</u>.
- Gardner C. S., Liu A. Z., Marsh D. R., Feng W. and Plane J. M. C. (2014) Inferring the
 global cosmic dust influx to the Earth's atmosphere from lidar observations of the
 vertical flux of mesospheric Na. J. Geophys. Res. Space Physics 119, 7870–7879.
- Genge M. J. (2008) Micrometeorites and their implications for meteors. *Earth, Moon, and Planets* 102, 525-535. <u>https://doi.org/10.1007/s11038-007-9185-z</u>.
- Genge M. J., Grady M. M. and Hutchison R. (1997) The textures and compositions of finegrained Antarctic micrometeorites: Implications for comparison with meteorites. *Geochim. Cosmochim. Acta* 61, 5149-5162. <u>https://doi.org/10.1016/S0016-</u>
 <u>7037(97)00308-6</u>.
- Genge M. J., Engrand C., Gounelle M. and Taylor S. (2008) The classification of
 micrometeorites. *Meteorit. Planet. Sci.* 43, 497-515. <u>https://doi.org/10.1111/j.1945-5100.2008.tb00668.x</u>

- Genge, M. J., van Ginneken M., Suttle M. D. and Harvey R.P. (2018) Accumulation
 mechanisms of micrometeorites in an ancient supraglacial moraine at Larkman
 Nunatak, Antarctica. *Meteorit. Planet. Sci.* 53, 2051-2066.
 https://doi.org/10.1111/maps.13107.
- Genge M. J., Van Ginneken M. and Suttle M. D. (2020) Micrometeorites: Insights into the
 flux, sources and atmospheric entry of extraterrestrial dust at Earth. *Planetary and Space Science* 187, #104900. <u>https://doi.org/10.1016/j.pss.2020.104900</u>.
- Gilmour J. D., Lyon I. C., Johnston W. A. and Turner G. (1994) RELAX: An ultrasensitive,
 resonance ionization mass spectrometer for xenon. *Rev. Sci. Instr.* 65, 617-625.
 <u>https://doi.org/10.1063/1.1145127</u>.
- Grün E., Zook H. A., Fechtig H. and Giese R. H. (1985) Collisional balance of the meteoritic
 complex. *Icarus* 62, 244–272. <u>https://doi.org/10.1016/0019-1035(85)90121-6</u>.
- Haack H., Grau T., Bischoff A., Horstmann M., Wasson J. T., Sørensen A., Laubenstein M.,
 Ott U., Palme H., Gellissen M., Greenwood R. C., Pearson V. K., Franchi I. A., Gabelica
 Z. and Schmitt-Kopplin P. (2012) Maribo-A new CM fall from Denmark. *Meteorit. Planet. Sci.* 47, 30–50. <u>https://doi.org/10.1111/j.1945-5100.2011.01311.x.</u>
- Heber V. S., Wieler R., Baur H., Olinger C., Friedmann T. A. and Burnett D. S. (2009) Noble
 gas composition of the solar wind as collected by the Genesis mission. *Geochim. Cosmochim. Acta* 73, 7414-7432. <u>https://doi.org/10.1016/j.gca.2009.09.013</u>.
- Heber V. S., Baur H., Bochsler P., McKeegan K. D., Neugebauer M., Reisenfeld D. B.,
 Wieler R. and Wiens R. C. (2012) Isotopic Mass Fractionation of Solar Wind: Evidence
 from fast and slow Solar Wind collected by the Genesis mission. *Astrophy. J.* 759, 121.
 <u>https://doi.org/10.1088/0004-637X/759/2/121</u>.
- Hébrard E. and Marty B. (2014) Coupled noble gas-hydrocarbon evolution of the early Earth
 atmosphere upon solar UV irradiation. *Earth Planet. Sci. Lett.* 385, 40-48.
 <u>https://doi.org/10.1016/j.epsl.2013.10.022</u>.
- Huss G. R. and Lewis R. S. (1994) Noble gases in presolar diamonds I: Three distinct
 components and their implications for diamond origins. *Meteoritics* 29, 791-810.
 <u>https://doi.org/10.1111/j.1945-5100.1994.tb01094.x.</u>
- Imae N., Taylor S. and Iwata N. (2013) Micrometeorite precursors: Clues from the
 mineralogy and petrology of their relict minerals. *Geochim. Cosmochim. Acta* 100, 116 157. <u>https://doi.org/10.1016/j.gca.2012.09.052</u>.
- Ishii H. A., Bradley J. P., Dai Z. R., Chi M., Kearsley A. T., Burchell M. J., Browning N. D.,
 and Molster F. (2008) Comparison of comet 81P/Wild 2 dust with interplanetary dust
 from comets. *Science* 319, 447–450. <u>https://doi.org/10.1126/science.1150683</u>.
- Kimura, H., Okamoto, H. and Mukai, T. (2002) Radiation pressure and the PoyntingRobertson effect for fluffy dust particles. *Icarus* 157, 349–361.
 <u>https://doi.org/10.1006/icar.2002.6849</u>.

- Kurat G., Koeberl C., Presper T., Brandstätter F. and Maurette M. (1994) Petrology and
 geochemistry of Antarctic micrometeorites. *Geochim. Cosmochim. Acta* 58, 3879-3904.
 <u>https://doi.org/10.1016/0016-7037(94)90369-7</u>
- 1167 Lee J. Y., Marti K., Severinghaus J. P., Kawamura K., Yoo H. S., Lee J. B. and Kim J. S.
- (2006) A redetermination of the isotopic abundances of atmospheric Ar. *Geochim. Cosmochim. Acta* 70, 4507-4512. <u>https://doi.org/10.1016/j.gca.2006.06.1563</u>.
- Love S. G and Brownlee D. E. (1993) A direct measurement of the terrestrial mass accretion
 rate of cosmic dust. *Science* 262, 550-553.
 https://doi.org/10.1126/science.262.5133.550.
- Marty B. (2012) The origins and concentrations of water, carbon, nitrogen and noble gases
 on Earth. *Earth Planet. Sci. Lett.* 313-314, 56-66.
 https://doi.org/10.1016/j.epsl.2011.10.040.
- Marty B., Matrajt G., Zimmermann L., Engrand C. and Duprat J. (2002) Nitrogen and noble
 gas isotopes in Antarctic micrometeorites. *Lunar Planet. Sci.* 33, abstract #1578.
- Marty B., Robert P. and Zimmermann L. (2005) Nitrogen and noble gases in
 micrometeorites. *Meteorit. Planet. Sci.* 40, 881-894. <u>https://doi.org/10.1111/j.1945-5100.2005.tb00161.x</u>.
- Marty B., Palma R. L., Pepin R. O., Zimmermann L., Schlutter D. J., Burnard P. G., Westphal
 A. J., Snead C. J., Bajt S., Becker R. H. and Simones J. E. (2008) Helium and neon
 abundances and compositions in cometary matter. *Science* 319, 75-78.
 <u>https://doi.org/10.1126/science.1148001</u>.
- Marty B., Avice G., Sano Y., Altwegg K., Balsiger H., Hässig M., Morbidelli A., Mousis O.
 and Rubin M. (2016) Origins of volatile elements (H, C, N, noble gases) on Earth and
 Mars in light of recent results from the ROSETTA cometary mission. *Earth Planet. Sci. Lett.* 441, 91-102. <u>https://doi.org/10.1016/j.epsl.2016.02.031</u>.
- Marty B., Altwegg K., Balsiger H., Bar-Nun A., Bekaert D. V., Berthelier J.-J., Bieler A.,
 Briois C., Calmonte U., Combi M., De Keyser J., Fiethe B., Fuselier S. A., Gasc S.,
 Gombosi T. I., Hansen K. C., Hässig M., Jäckel A., Kopp E., Korth A., Le Roy L., Mall
 U., Mousis O., Owen T., Rème H., Rubin M., Sémon T., Tzou C.-Y., Waite J. H. and
 Wurz P. (2017) Xenon isotopes in 67P/Churyumov-Gerasimenko show that comets
 contributed to Earth's atmosphere. *Science* 356, 1069-1072.
 https://doi.org/10.1126/science.aal3496.
- Maurette M. (2006) Micrometeorites and the Mysteries of Our Origins. Springer-Verlag,
 Berlin Heidelberg, New York, 330 pp..
- Maurette M., Olinger C., Christophe Michel-Levy M., Kurat G., Pourchet M., Brandstätter F.
 and Bourot-Denise M. (1991) A collection of diverse micrometeorites recovered from
 100 tonnes of Antarctic blue ice. *Nature* 351, 44-47. <u>https://doi.org/10.1038/351044a0</u>.

- Maurette M., Duprat J., Engrand C., Gounelle M., Kurat G., Matrajt G. and Toppani A.
 (2000) Accretion of neon, organics, CO₂, nitrogen and water from large interplanetary
 dust particles on the early Earth. *Planet. Space Sci.* 48, 1117-1137.
 https://doi.org/10.1016/S0032-0633(00)00086-6.
- Meshik A., Hohenberg C., Pravdivtseva O. and Burnett D. (2014) Heavy noble gases in solar
 wind delivered by Genesis mission. *Geochim. Cosmochim. Acta* 127, 326-347.
 https://doi.org/10.1016/j.gca.2013.11.030.
- Meshik A. P., Pravdivtseva O. V., Hohenberg C. M. and Burnett D. S. (2015) Refined
 composition of solar wind xenon delivered by Genesis: Implication for primitive
 terrestrial xenon. *Lunar Planet Sci.* 46, #2640 (abstract).
- Messenger S., Keller L. P., Stadermann F. J., Walker R. M. and Zinner E. (2003) Samples of stars beyond the solar system: silicate grains in interplanetary dust. *Science* 300, 105– 108. <u>https://doi.org/10.1126/science.1080576</u>.
- Nagao K., Okazaki R., Nakamura T., Miura Y. N., Osawa T., Bajo K.-i., Matsuda S., Ebihara
 M., Ireland T. R., Kitajima F., Naraoka H., Noguchi T., Tsuchiyama A., Yurimoto H.,
 Zolensky M. E., Uesugi M., Shirai K., Abe M., Yada T., Ishibashi Y., Fujimura A.,
 Mukai T., Ueno M., Okada T., Yoshikawa M. and Kawaguchi J. (2011) Irradiation
 history of Itokawa regolith material deduced from noble gases in the Hayabusa samples. *Science* 333, 1128-1131. https://doi.org/10.1126/science.1207785.
- Nakamura T., Noguchi T., Ozono Y., Osawa T. and Nagao K. (2005) Mineralogy of
 ultracarbonaceous large micrometeorites. 68th Annu. Meteorit. Soc. Meeting, abstract
 #5046.
- Nesvorný D., Jenniskens P., Levison H. F., Bottke W. F., Vokrouhlický D. and Gounelle M.
 (2010) Cometary origin of the Zodiacal Cloud and carbonaceous micrometeorites.
 Implications for hot debris disks. *Astrophys. Journ.* 713, 816-836.
- 1226 <u>https://doi.org/10.1088/0004-637X/713/2/816</u>.
- Nier A. O. and Schlutter D. J. (1990) Helium and neon isotopes in stratospheric particles.
 Meteoritics 25, 263–267. <u>https://doi.org/10.1111/j.1945-5100.1990.tb00710.x</u>.
- Nier A. O. and Schlutter D. J. (1993) The thermal history of interplanetary dust particles
 collected in the Earth's stratosphere. *Meteoritics* 28, 675-681.
 https://doi.org/10.1111/j.1945-5100.1993.tb00639.x.
- Noguchi T., Ohashi N., Tsujimoto S., Mitsunari T., Bradley J. P., Nakamura T., Toh S.,
 Stephan T., Iwatag N., and Imae N. (2015) Cometary dust in Antarctic ice and snow:
 Past and present chondritic porous micrometeorites preserved on the Earth's surface. *Earth Planet. Sci. Lett.* 410, 1-11. <u>https://doi.org/10.1016/j.epsl.2014.11.012</u>.
- Ogliore R. C., Palma R. L., Stodolna, J., Nagashima K., Pepin R. O., Schlutter D. J.,
 Gainsforth Z., Westphal A. J. and Huss G. R. (2020) Q-Gases in a late-forming
 refractory interplanetary dust particle: A link to comet Wild 2. *Geochim. Cosmochim. Acta* 271, 116-131. <u>https://doi.org/10.1016/j.gca.2019.11.033</u>.

- Okazaki R., Noguchi T., Tsujimoto S.-i., Tobimatsu Y., Nakamura T., Ebihara M., Itoh S.,
 Nagahara H., Tachibana S., Terada K. and Yabuta H. (2015) Mineralogy and noble gas
 isotopes of micrometeorites collected from Antarctic snow. *Earth, Planets and Space* 67,
 90. https://doi.org/10.1186/s40623-015-0261-8.
- Olinger C. T., Maurette M., Das J. P. and Meshik A. (2013) Noble gas contents of unmelted
 Cap-Prudhomme "Giant Micrometeorites". *Lunar Planet. Sci.* 44, abstract #2278.
- Osawa T. and Nagao K. (2002a) Noble gas compositions of Antarctic micrometeorites
 collected at the Dome Fuji station in 1996 and 1997. *Meteorit. Planet. Sci.* 37, 911–936.
 https://doi.org/10.1111/j.1945-5100.2002.tb00867.x.
- Osawa T. and Nagao K. (2002b) On low noble gas concentrations in Antarctic
 micrometeorites collected from Kuwagata Nunatak in the Yamato meteorite ice field.
 Antarct. Meteorite Res. 15, 165-177.
- Osawa T., Nagao K., Nakamura T. and Takaoka N. (2000) Noble gas measurement in
 individual micrometeorites using laser gas-extraction system. *Antarct. Meteorite Res.* 13,
 322-341.
- Osawa T., Nagao K., Noguchi T., Nakazawa A. and Mikada J. I. (2003a) Remnant
 extraterrestrial noble gases in Antarctic cosmic spherules. *Antarct. Meteorite Res.* 16, 196-219.
- Osawa T., Nakamura T. and Nagao K. (2003b) Noble gas isotopes and mineral assemblages
 of Antarctic micrometeorites collected at the meteorite ice field around the Yamato
 Mountains. *Meteorit. Planet. Sci.* 38, 1627–1640. <u>https://doi.org/10.1111/j.1945-</u>
 <u>5100.2003.tb00005.x</u>.
- Osawa T., Yamamoto Y., Noguchi T., Iose A. and Nagao K. (2010) Interior textures,
 chemical compositions, and noble gas signatures of Antarctic cosmic spherules: Possible
 sources of spherules with long exposure ages. *Meteorit. Planet. Sci.* 45, 1320-1339.
 <u>https://doi.org/10.1111/j.1945-5100.2010.01093.x.</u>
- Ott U. (2014) Planetary and pre-solar noble gases in meteorites. *Chemie der Erde* –
 Geochem. 74, 519-544. <u>https://doi.org/10.1016/j.chemer.2014.01.003</u>.
- Ott U. and Begemann F. (1985) Are all the Martian meteorites from Mars? *Nature* 317, 509 512. <u>https://doi.org/10.1038/317509a0</u>.
- Ott U., Heck P. R., Gyngard F., Wieler R., Wrobel F., Amari S. and Zinner E. (2009) He and
 Ne ages of large presolar silicon carbide grains: Solving the recoil problem. *Pub. Astronom. Soc. Australia* 26, 297–302. https://doi:10.1071/AS08039.
- Ott U., Swindle T. D. and Schwenzer S. P. (2019) Noble gases in Martian meteorites:
 Budget, sources, sinks, and processes, In: *Volatiles in the Martian Crust* (ed. J. Filiberto and S.P. Schwenzer), 35-70, Elsevier.

- Owen T. (2008) The contributions of comets to planets, atmospheres, and life: Insights from
 Cassini-Huygens, Galileo, Giotto and inner planet missions. *Space Sci. Rev.* 138, 301 316. https://doi.org/10.1007/s11214-008-9306-7.
- 1279 Owen T. and Bar-Nun A. (1993) Noble gases in atmospheres. *Nature* **361**, 693-694.
- Owen T., Bar-Nun A. and Kleinfeld I. (1992) Possible cometary origin of heavy noble gases
 in the atmospheres of Venus, Earth and Mars. *Nature* 358, 43-46.
 https://doi.org/10.1038/358043a0.
- Ozima M. and Podosek F. A. (2002) Noble Gas Geochemistry. Cambridge University Press,
 Cambridge, New York, Oakleigh, Madrid, Cape Town. 286 pp..
- Pepin R. O. (1991) On the origin and early evolution of terrestrial planet atmospheres and
 meteoritic volatiles. *Icarus* 92, 2-79. <u>https://doi.org/10.1016/0019-1035(91)90036-S</u>.
- Pepin R. O. (2006) Atmospheres on the terrestrial planets: Clues to origin and evolution.
 Earth Planet. Sci. Lett. 252, 1-14. <u>https://doi.org/10.1016/j.epsl.2006.09.014</u>.
- Pepin R. O. and Porcelli D. (2002) Origin of noble gases in the terrestrial planets. In: *Noble Gases in Geochemistry and Cosmochemistry* (eds. D. Porcelli, C. J. Ballentine and R. Wieler). *Rev. Min. Geochem.* 47, 191-246. <u>https://doi.org/10.2138/rmg.2002.47.7</u>.
- Pepin R. O., Palma R. L. and Schlutter D. J. (2000) Noble gases in interplanetary dust
 particles, I: The excess helium-3 problem and estimates of the relative fluxes of solar
 wind and solar energetic particles in interplanetary space. *Meteorit. Planet. Sci.* 35, 495504. <u>https://doi.org/10.1111/j.1945-5100.2000.tb01431.x</u>.
- Pepin R. O., Palma R. L. and Schlutter D. J. (2001) Noble gases in interplanetary dust
 particles, II: Excess helium-3 in cluster particles and modeling constraints on
 interplanetary dust particle exposure to cosmic-ray irradiation. *Meteorit. Planet. Sci.* 36,
 1515-1534. <u>https://doi.org/10.1111/j.1945-5100.2001.tb01843.x</u>.
- Pernet-Fisher J. F., Joy K. H. and Gilmour J. D. (2020) Identifying primitive noble gas
 components in lunar ferroan anorthosites. *Icarus* 352, 113977.
 https://doi.org/10.1016/j.icarus.2020.113977.
- Piani L., Marrocchi Y., Rigaudier T., Vacher L. G., Thomassin D. and Marty B. (2020)
 Earth's water may have been inherited from material similar to enstatite chondrite
 meteorites. *Science* 369, 1110-1113. https://doi.org/10.1126/science.aba1948.
- Rochette P., Folco L., Suavet C., van Ginneken M., Gattacceca J., Perchiazzi N., Braucher R.
 and Harvey R. P. (2008) Micrometeorites from the Transantarctic Mountains. *Proc. National Acad. Sci.* 105, 18206-18211. <u>https://doi.org/10.1073/pnas.0806049105</u>.
- Rojas J., Duprat J., Engrand C., Dartois E., Delauche L., Godard M., Gounelle M., CarrilloSánchez J. D., Pokorný P. and Plane J. M. C. (2021) The micrometeorite flux at Dome
 C (Antarctica), monitoring the accretion of extraterrestrial dust on Earth. *Earth Planet. Sci. Lett.* 560, # 116794. https://doi.org/10.1016/j.epsl.2021.116794.

- Rubin M., Altwegg K., Balsiger H., Bar-Nun A., Berthelier J.-J., Briois C., Calmonte U., 1313 Combi M., De Keyser J., Fiethe B., Fuselier S. A., Gasc S., Gombosi T. I., Hansen K. 1314 C., Kopp E., Korth A., Laufer D., Le Roy L., Mall U., Marty B., Mousis O., Owen T., 1315 Rème H., Sémon T., Tzou C.-Y., Waite J. H. and Wurz P. (2018) Krypton isotopes and 1316 noble gas abundances in the coma of 67/PChuryumov-Gerasimenko. Science Advances 1317 1318 4, eaar6297. https://doi.org/10.1126/sciadv.aar6297. Sarda Ph., Staudacher Th. and Allègre C. J. (1991) Complete rare gas study of a very large 1319 unmelted cosmic dust particle from Greenland. Lunar Planet. Sci. 22, 1165-1166 1320 (abstract). 1321
- Schmitz B., Feist R., Meier M. M. M., Martin E., Heck P. R, Lenaz D., Topa D., Busemann
 H., Maden C., Plant A. A. and Terfelt F. (2019) The micrometeorite flux to Earth
 during the Frasnian–Famennian transition reconstructed in the Coumiac GSSP section,
 France. *Earth Planet. Sci. Lett.* **522**, 234-243.
 https://doi.org/10.1016/j.epsl.2019.06.025.
- Schwenzer S. P., Herrmann S., Mohapatra R. K. and Ott U. (2007) Noble gases in mineral
 separates from three shergottites: Shergotty, Zagami, and EETA79001. *Meteorit*.
- 1329 Planet. Sci. 42, 387-412. <u>https://doi.org/10.1111/j.1945-5100.2007.tb00241.x</u>.
- Shyam Prasad M., Rudraswami N. G., de Araujo A. A. and Khedekar V. D. (2018)
 Characterisation, sources and flux of unmelted micrometeorites on Earth during the last
 ~50,000 Years. *Scientific Reports* 8, 8887. <u>https://doi.org/10.1038/s41598-018-27158-x</u>.
- Spring N. H. (2014) Investigating the Origin and Distribution of Primordial Noble Gases and
 Volatiles in Asteroids and Comets: A Comparative Study of Interplanetary Dust Particles
 (IDPs), Antarctic Micrometeorites (AMMs) and Carbonaceous Chondrites. PhD Thesis. *University of Manchester, United Kingdom*, 178 p.
- Stuart F., Harrop P. J., Knott S., and Turner G. (1999) Laser extraction of helium isotopes
 from Antarctic micrometeorites: Source of He and implications for the flux of
 extraterrestrial ³He to earth. *Geochim. Cosmochim. Acta* 63, 2653-2665.
 https://doi.org/10.1016/S0016-7037(99)00161-1.
- Suavet C., Cordier C., Rochette P., Folco L., Gattacceca J., Sonzogni C. and Damphoffer D.
 (2011) Ordinary chondrite-related giant (> 800µm) cosmic spherules from the
 Transantarctic Mountains, Antarctica. *Geochim. Cosmochim. Acta* 75, 6200-6210.
 <u>https://doi.org/10.1016/j.gca.2011.07.034</u>.
- Suttle, M. D. and Folco L. (2020) The extraterrestrial dust flux: Size distribution and mass
 contribution estimates inferred from the TransantarcticMountains (TAM)
 micrometeorite collection. *Journal of Geophysical Research: Planets* 125,
 e2019JE006241. https://doi.org/10.1029/2019JE006241.
- Taylor S., Lever J. H. and Harvey R. P. (2000) Numbers, types, and compositions of an
 unbiased collection of cosmic spherules. *Meteorit. Planet. Sci.* 35, 651-666.
 <u>https://doi.org/10.1111/j.1945-5100.2000.tb01450.x.</u>

- Taylor S., Herzog G. F. and Delaney J. S. (2007) Crumbs from the crust of Vesta:
 Achondritic cosmic spherules from the South Pole water well. *Meteorit.Planet. Sci.*42, 223-233. <u>https://doi.org/10.1111/j.1945-5100.2007.tb00229.x.</u>
- Taylor S., Matrajt G. and Guan Y. (2012) Fine-grained precursors dominate the
 micrometeorite flux. *Meteorit. Planet. Sci.* 47, 550-564. <u>https://doi.org/10.1111/j.1945-</u>
 <u>5100.2011.01292.x.</u>
- Toppani A., Libourel G., Engrand C. and Maurette M. (2001) Experimental simulation of
 atmospheric entry of micrometeorites. *Meteorit. Planet. Sci.* 36, 1377-1396.
 https://doi.org/10.1111/j.1945-5100.2001.tb01831.x.
- Trappitsch R. and Leya I. (2013) Cosmogenic production rates and recoil loss effects in
 micrometeorites and interplanetary dust particles. *Meteorit. Planet. Sci.* 48, 195-210.
 <u>https://doi.org/10.1111/maps.12051</u>.
- Trappitsch R. and Leya I. (2016) Production and recoil loss of cosmogenic nuclides in presolar grains. *Astrophy. J.* 823, 12. <u>https://doi.org/10.3847/0004-637X/823/1/12</u>.
- Trieloff M., Kunz J., Clague D. A., Harrison D. and Allègre C. J. (2000) The nature of
 pristine noble gases in mantle plumes. *Science* 288, 1036-1038.
 <u>https://doi.org/10.1126/science.288.5468.1036</u>.
- van Ginneken M., Folco L., Cordier C. and Rochette P. (2012) Chondritic micrometeorites
 from the Transantarctic Mountains. *Meteorit. Planet. Sci.* 47, 228-247.
 <u>https://doi.org/10.1111/j.1945-5100.2011.01322.x.</u>
- 1372 Vogel N., Heber V. S., Baur H., Burnett D. S. and Wieler R. (2011) Argon, krypton, and
 1373 xenon in the bulk solar wind as collected by the Genesis mission. *Geochim. Cosmochim.* 1374 Acta, 75, 3057-3071. <u>https://doi.org/10.1016/j.gca.2011.02.039</u>.
- 1375 Vogt M., Hopp J., Gail H.-P., Ott U. and Trieloff M. (2019) Acquisition of terrestrial neon
 1376 during accretion A mixture of solar wind and planetary components. *Geochim.*1377 *Cosmochim. Acta* 264, 141-164. <u>https://doi.org/10.1016/j.gca.2019.08.016</u>.
- Wieler R. (2002a) Noble gases in the Solar System. In: *Noble Gases in Geochemistry and Cosmochemistry* (eds. D. Porcelli, C. J. Ballentine and R. Wieler). *Rev. Min. Geochem.*47, 21-70. <u>https://doi.org/10.2138/rmg.2002.47.2</u>.
- Wieler R. (2002b) Cosmic-ray-produced noble gases in meteorites. In: *Noble Gases in Geochemistry and Cosmochemistry* (eds. D. Porcelli, C. J. Ballentine and R. Wieler). *Rev. Min. Geochem.* 47, 125-170. <u>https://doi.org/10.2138/rmg.2002.47.5</u>.
- Wieler R., Grimberg A. and Heber V. S. (2007) Consequences of the non-existence of the
 "SEP" component for noble gas geo-and cosmochemistry. *Chem. Geol.* 244, 382-390.
 <u>https://doi.org/10.1016/j.chemgeo.2007.06.026</u>.
- Wyatt S. P. and Whipple F. L. (1950) The Poynting-Robertson effect on meteor orbits.
 Astrophys. Journ. 111, 134-141. https://doi.org/ <u>10.1086/145244</u>.

1389	Yabuta, H., Noguchi T., Itoh S., Nakamura T., Miyake A., Tsujimoto S., Ohashi N.,
1390	Sakamoto N., Hashiguchi M., Abe Ki., Okubo A., Kilcoyne A. L. D., Tachibana S.,
1391	Okazaki R., Terada K., Ebihara M. and Nagahara H. (2017) Formation of an
1392	ultracarbonaceous Antarctic micrometeorite through minimal aqueous alteration in a
1393	small porous icy body. <u>Geochim. Cosmochim. Acta</u> 214, 172-190.
1394	https://doi.org/10.1016/j.gca.2017.06.047.