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The Exploration of Neptune: A Noble Gas and Volatile Perspective

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Abstract：Most of the probes visiting other bodies in our Solar system only focused，due to technical shortcomings，on the exploration of the closer planets and planetary bodies and/or their natural satellites，i.e. Mercury，Venus，the Moon，Mars，and Jupiter，the comet 67P/Churyumov-Gerasimenko，or the 25143 Itokawa near-Earth asteroid. At present time，no specific missions to one of the two ice giants of our Solar System，Uranus and Neptune，has been planned. Our knowledge of Uranus and Neptune is，therefore，so far restricted to the data which have been collected during the flyby of the Voyager 2 mission，in January 1986 and August 1989，respectively，and to observations with the Hubble Space Telescope and the Keck Telescope. Ice giants are，in our galaxy，thought to be much more abundant than gas giant planets such as Jupiter or Saturn，therefore a better knowledge of ice giants is essential for our understanding of exoplanet candidates. Among other scientific goals，the atmospheric composition of ice giants，with a particular emphasis on their noble gas and volatile distribution，is of great significance，and can constrain models about their formation and evolution. In this review，we report，in a first part，the volatile inventories and the measurements in the planetary bodies of our Solar System； in a second part， we will discuss the scientific background about the concentration，distribution，and evolution of noble gases and volatiles in Uranus and Neptune，and finally describe a possible scenario of a future interstellar probe visiting one of the two ice giants as well as the feasibility of such a space mission，in term of payloads selection and mission profile. We will as well briefly evoke the possibility of using an ion trap mass spectrometer，a potential payload for the ice giant atmospheric exploration，onboard a Chinese interstellar mission to the outer Solar system.

Keywords：ice giants；Neptune；volatiles；noble gases；isotopic ratios；mass spectrometry；entry probe

Highlights：

- The number of ice giant-mass detected exoplanets increased intensely in the past decades.
- The measurements of noble gases，volatiles and their isotopic ratios in ice giant planets would provide crucial information to better constrain their formation mechanisms.
- Mass spectrometry onboard an interstellar mission to Neptune can address these questions.

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Introduction

Most of the probes visiting other bodies in our Solar system only focused，due to technical challenges，on the exploration of the closer planets，including Mercury（i.e. Mariner 10，Messenger[1]；or BepiColombo scheduled for orbit insertion in 2025[2]），Venus（i.e. the past Venera and Vega missions，and ongoing[3]），the Moon（e.g，Apollo，NASA，USSR Luna program，Chang-E' series of missions[4-6] etc.），Mars（e.g，Sojourner，Spirit，Opportunity，Curiosity，and many

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more others scheduled），and Jupiter（i.e. Galileo，Juno[7]），and bodies，such as e.g，the comet 67P/Churyumov-Gerasimenko[8] or the near-Earth asteroid Itokawa[9]. However，our knowledge of the two ice giant planets Uranus and Neptune is only restricted to the data which have been collected during the flyby of the Voyager 2 mission，in January 1986 and August 1989，respectively，and to observations with the Hubble Space Telescope and the Keck Telescope. Ice giants therefore represent a challenge，both in terms of scientific feasibility and scientific knowledge. The increasing number of scientific articles and reviews recently published about the ice giants，their properties，and their exploration is a good indicator of the growing interest of the planetary science community for those so far unknown territories of the Solar system. Among them，we can cite the following Mousis et al，Jarmak et al，Atreya et al.，Hofstadter et al.，Mandt et al.，Mousis et al.，Sayanagi et al.，Vorburger et al.[10-18]，etc. One of the reasons for such a rapid increase in ice giant-related papers lies in the fact that the number of ice giant mass detected exoplanets increased intensely in the past decades[19]. Ice giants are indeed，in our galaxy，thought to be much more abundant than gas giant planets such as Jupiter or Saturn[20]. Among the 4 000 confirmed exoplanets detected up to January 2020，about 51% have the size of Neptune（i.e. 1.7～6 Earth radius，R_E），whereas only 14% are “Jupiter-like”（i.e. with radius of 6～15R_E），18% have “Super Earth size”（i.e. 1.25～1.7 R_E）[13]. Consequently，a better knowledge of ice giants is essential for our understanding of exoplanet candidates. Little is known about the formation conditions of ice giant planets，though it is believed that they must have formed quickly before the dissipation of the protoplanetary disk（between ～3～10 million years[21]）. Core accretion or disk instability are the two mechanisms commonly evoked to explain the ice giant’s formation[22]. However，the core accretion model scenario still cannot fully explain the formation of our ice giants，or at least，their formation mechanisms are not yet completely understood[23]. Given this fact，the chemical composition of Uranus and Neptune is therefore believed to represent the environment of the proto-Sun and the proto-planetary disc；knowing their chemical compositions give access to the formation and evolution of our Solar System[11，18]. In addition，their actual position in our Solar System is incompatible with the amount of material available to build such large planets；with a mass of $1.024\,3 \times 10^{26}$ kg，Neptune’s mass represents 17 times the mass of the Earth. A model predicted that the ice-giants might have formed in a region of space much closer to the proto-Jupiter，and then migrated in their actual orbits when Jupiter acquired its final mass[24]. The Nice model[25-27] predicts that the orbits of Uranus and Neptune shifted early after the formation of the Solar System. As confirmed by Desch[28]，it is now assumed that Neptune formed closer to the Sun than Uranus，around 10 AU and 20 AU，respectively，and then both planets switched their orbits ～600 Ma after the formation of the Solar System. The mechanisms of their formation itself is not fully constrained，as even inducing small changes in the initial input parameters in the models would contribute to differences in the final mass and solid-to-gas ratios[29]. Noble gases（He，Ne，Ar，Kr，and Xe），and volatiles，among others，as well as their isotopic ratios，provide constraints for our understanding of the formation processes and the evolution of planetary bodies. Precise measurements of their abundances and accurate determination of their isotopic ratios are therefore necessary and represent a primary target in planetary atmosphere exploration.

1　The noble gas and volatile inventories as investigated by probes in the Solar system and in the returned samples in laboratory

In the following chapter，we will briefly review the results about noble gas and volatiles inventories in some of the planetary bodies of the Solar System，investigated by probes and，if available，by sample return in laboratories（Moon and Mars with Martian meteorites）. It is not here our intention to give a comprehensive and detailed review about the volatile abundances in the Solar System，but to only mention the set of data so far available，and to shortly discuss the state-of-the art of volatile measurements in extraterrestrial bodies（section 3.2. will as well discuss this point）.

1.1　The inner Solar system：Mercury，Venus，the Moon and Mars

1.1.1　The Moon

The Earth’s Moon is one of the most investigated bodies in our Solar System. The Moon，particularly，has been widely studied and explored by manned and unmanned missions during the past decades. It is in fact，up to date，the only planetary body for which samples have been collected and brought back to Earth，either by human or robotic missions[30]. The Moon programs，both from the USA and Soviet Union space agencies，were initiated in the late 1950s. In total，between 1969 and 1972，six *Apollo* missions brought back 382 kilograms of lunar samples consisting of rocks，core samples，pebbles，sand，and dust. The six space flights returned 2 200 separate samples from six different exploration sites on the Moon. Besides，the three automated USSR spacecraft returned ～300 grams of lunar samples. In addition to the -so far- only lunar samples available after the US and Soviet missions to the Moon，lunar meteorites excavated from the Moon surface，and or interior by major impacts，represent probably the best and easily accessible samples to study the Moon. At the time we are writing this review，a total of 415 lunar meteorites are officially approved by the Meteoritical Society（see Meteoritical Bulletin database as of August 2，2020）. Almost 2/3 of these lunar meteorites，all finds，have been collected in North and/or Northwest Africa（NWA），others come from the Arabian Peninsula or Antarctica[31]. Although the Earth's Moon has been considered as being volatile-depleted[32-33]，lunar meteorites contain volatiles，and，among others noble gases（He，Ne，Ar，Kr，and Xe）. The latter are produced by a constant bombardment of the Moon by energetic particles：（i）solar wind（SW）implanted particles；（ii）production of cosmogenic radionuclides on the lunar regolith by the interaction of energetic particles，solar cosmic-rays and galactic cosmic-rays（SCRs and GCRs，respectively[34-38]）. In addition，noble gases are produced in-situ by the radioactive decay of elements or can be delivered to the Moon by asteroids or comets[39-40]. The possibility to have access to lunar samples，e.g，rocks or soil brought back by the *Apollo* and *Luna* missions，or lunar meteorites，initiated a new era in lunar research. Of particular scientific interest is the investigation of the evolution of the Moon and its surface through time，or eventually the distribution of volatiles and other mineralogical resources for future potential In Situ Resource Utilization（ISRU）applications[41-42]. In a recent review，Tartèse et al.[30] summarized the most important questions which sample return would address. The volatile inventories，including noble gases，have scientific significance regarding e.g，the lunar volcanism，the structure and composition of the lunar interior，the composition and distribution of lunar volatiles at the pole，the GCR and SW information recorded in the lunar surface samples，or the long timescale cosmic-ray energy spectrum reconstruction.

The noble gas abundances at the Moon have been measured by mass spectrometry，mainly. It is indeed the most important analytical technique used to quantify noble gas concentrations and their isotopic ratios，both in space in meteorites and/or returned samples in the laboratory. For example，the He and Ar abundances in the lunar exosphere have been quantified by ion- and neutral mass spectrometry，Ar-Ar ages，or exposure ages，SW component evaluation，have been performed by mass spectrometry in the laboratory，and additional information about Kr and Xe concentrations have been measured from orbit by UV spectroscopy[38].

In order to primarily investigate the abundances of hydrogen detected at the polar regions of the Moon，the Lunar Crater Observation and Sensing Satellite（LCROSS，October 2009）aimed at sending a projectile into a crater，provoking a giant plume of debris，and to measure the composition of the material released by the impact thanks to a spacecraft. Doing so，the amount of water trapped as ices in a permanently shadowed crater near the lunar polar region was further explored. The LCROSS science payload consists of two near-infrared spectrometers，a visible light spectrometer，two mid-infrared cameras，two near-infrared cameras，a visible camera，and a visible radiometer[43]. The ejecta was mainly composed of water（77%），H_2S（13%），NH_3（5%），C_2H_4（2%），CO_2（2%），and CH_3OH（1%）[44].

1.1.2 Mercury

The volatile inventories at Mercury have been essentially obtained by the Mercury Surface，Space Environment，Geochemistry and Ranging（Messenger）spacecraft；Mercury was first believed to be severely depleted in volatiles because being the closest planet of the Sun，but the first missions，like the Mariner 10 flybys，coupled with ground-based observations revealed the presence of exospheric H，He，and O and the presence of polar volatiles in radar-reflective deposits[45]. The Messenger mission truly revealed that Mercury is a volatile-rich planet；with its embarked payloads including，among others，X-Ray spectrometer，Gamma-ray spectrometer，or Neutron spectrometer，Messenger returned crucial information about the abundances and distribution of volatiles on the surface of the planet. The concentrations in e.g，C，Na，S，Cl，and K have been measured and suggested that Mercury possesses high levels of indigenous volatiles（Greenwood et al.[45]，and references herein）. However，the data obtained by Mariner and Messenger only reveal the abundances of surface volatiles；abundances of those magmatic volatiles in the interior of Mercury have been assumed from the probe data，and linked to volcanic units in e.g，the North polar region of the planet. More details are presented in Greenwood et al.[45].

1.1.3　Venus

Unlike Mars（see next section）and the Earth's Moon，there are no meteorites from Venus so far，therefore no indirect access to the Venusian atmosphere. Venus has been studied through the USSR Venera suite of missions，Venera 4（1967）being the first probe to enter another planet's atmosphere and to return data. Later，Venera 7（1970）was the first successful spacecraft landing on another planet and the first transmission from another planet's surface. Venera 7 only survived 23 minutes in the harsh environment of the Venusian atmosphere. These missions and the later Pioneer suite of missions revealed that CO_2 and N_2 are the major constituents of the atmosphere，together with SO_2，220 times higher than the S abundance in the Earth. The other volatiles are at ppm and ppb levels，and noble gases are as follows：^{4}He = 0.6～12 ppm；Ne ～ 7 ppm；^{38}Ar = 5.5 ppm；^{84}Kr = 25 ppb；$^{129,\ 132}$Xe < 10 ppb[38，46]. Isotopic ratios have been measured either by mass spectrometers or Infra-Red spectrometers embarked onboard Pioneer and Venera（Fegley，2014[46]，and references therein）. Crucial information，such as the D/H ratio and the noble gas isotopic ratios have been collected. The D/H ratio at Venus is ～150 times greater than on Earth（SMOW），which suggests that Venus had initially more water which was subsequently lost.

1.1.4　Mars

Mars is，after the Earth's Moon，one of the most studied bodies of the inner Solar System，thanks to designated probes and through measurements of Martian meteorites in the laboratory. Indeed，Martian meteorites represent the only samples from Mars which are available so far. Up to date，there are 276 records for Martian meteorites in the see Meteoritical Bulletin database as of August 2，2020.

During the past decades，several space missions to Mars measured the atmospheric compounds of the atmosphere，and，among others，the noble gas abundance，as well as their isotopic ratios by means of mass spectrometry or EUV spectroscopy. Among those missions，one can mention Viking suites of missions（1976—1982），Curiosity（2012），the Mars Atmosphere and Volatile Evolution（MAVEN），or the Mars Orbiter Mission（MOM）. More details about those Martian missions can be found in e.g，Owen and Biemann（1976），Owen et al.，or Mahaffy et al.[47-50]，and in section 3.2. of this paper. Noble gases found at Mars and in Martian meteorite generally contain components such as cosmogenic，trapped，radiogenic，and fissiogenic components，which can be used as potential tools to understand the Martian atmospheric loss processes over geological time，or as clues on volatile reservoirs and transport processes on Mars. Volatiles at Mars（S，Cl，C，H_2O，etc.）have been measured by e.g，the instruments onboard the Phoenix lander（2008），at the Northern plains of Mars. Among those instruments，the Thermal and Evolved Gas Analyzer（TEGA）consisted of scanning calorimetry cells coupled to a magnetic-sector mass spectrometer. For more details，see Kounaves and Oberlin，2018[51]，as well as references therein.

1.2　The outer Solar system：Jupiter and Saturn's moon Titan

The two gas giants Jupiter and Saturn and their satellites have been explored through e.g，the Galileo mission（Jupiter）or Cassini-Huygens（Saturn's moon Titan）.

The Galileo Probe Mass Spectrometer（GPMS）which investigated Jupiter in 1995 and revealed the composition of the Jovian atmosphere. The GPMS，as described in e.g，Niemann et al.（1992 & 1996）[52-53]，has been designed to measure the abundances and isotopic ratios of minor and major species. Briefly，the Jovian atmosphere was inlet into the ion source of a quadrupole mass spectrometer. The mass spectrometer possesses a noble gas purification cell and two sample enrichment cells. The inlet units opened in sequence，at different depths；the incoming gas goes through two glass capillaries which diameter is ranging from 1.5～6 μm，well designed to reach a pressure lower than 10^{-4} mbar. After the gas passes through the capillaries，it penetrates the enrichment cells，equipped with a series of four getters which helped to reduce the abundances of hydrogen and reactive species，which could，otherwise，compromise or decrease the accuracy of the measurements：during their activation，reactive gases are reduced or eliminated，therefore allowing suitable and accurate measurements of noble gases. Hydrocarbons are further adsorbed in the enrichment cells. The adsorbed gases can be subsequently released by heating cycles while the Galileo Probe descends and flow towards the ion source. Hydrocarbons can then be enriched by factors of 100～500（depending on their mass），and by factors of 10～100 for e.g，Kr and Xe. The Galileo mission was as well equipped with a Helium Abundance Detector（HAD），a refractometer. Although helium abundances and isotopic ratio can be measured by mass spectrometers，HAD’s main objective was to perform precise measurements，at the percent level，of the He abundance in the Jovian atmosphere between 3～8 bars. Such accuracy in the measurements are necessary in order to precisely define the small differences between the He/H_2 abundances at Jupiter compared to the Sun. The He mole fraction measured in the Jovian troposphere by the HAD was found to be 0.135 9 ± 0.002 7，in really good agreement with the GPMS[54-55].

The ESA’s Huygens probe was part of the Cassini mission designed to explore Saturn. Its particular design aimed at protecting its suites of instruments during the 2 hrs and 27 minutes descent through the atmosphere of Saturn’s giant moon Titan. The Huygens probe successfully landed on Titan at about 11:30 UTC on January 14，2005. The Descent Module was equipped with a suite of scientific instruments and three different parachutes that were deployed in sequence to control its descent. The probe survived approximately 72 minutes on the surface of Titan. This was the first，and，so far，the only，landing of a probe in the outer solar system.

The probe was equipped with 6 different instruments，including，among others，the Huygens Atmospheric Structure Instrument（HASI），which measured the physical and electrical properties of the Titan atmosphere，a doppler wind experiment，to measure the wind speed，a descent imager/spectral radiometer which aimed at measuring the radiation balance in the atmosphere，or a Gas Chromatograph Mass Spectrometer（GCMS），for the measurements of the chemical compositions of atmospheric gases and aerosols. It consisted in the association of a gas sample inlet and enrichment systems，and a mass spectrometer coupled with a gas chromatograph in order to increase the measurement capability of the instrument by performing batch sampling during the descent in the atmosphere，and therefore to allow separation of the different species having different chemical properties for the subsequent measurement by the mass spectrometer. A full description of the mission scenario and the suite of payloads onboard the Huygens probe can be found in Niemann et al. or Meltzer[56-57].

2 The ice giants composition，from their interiors to the distribution of volatiles

2.1 The structure and composition of the atmosphere

2.1.1 The ice giants’ internal structure

As mentioned，ice giants’ internal structures like Uranus and Neptune differ from the gas giants’ interiors. Whereas Jupiter and Saturn are mainly subdivided into three main layers composed of an outer hydrogen envelope，an inner hydrogen-helium envelope，enriched in He，and a central dense core，Uranus and Neptune are mainly composed of “ices”（see section 3.1.3. for more details about the cloud layer structure）of e.g，water，methane，or ammonia，which could become ionic fluid or oceans with great depths. The diagrams in Figure 1 represent the interior of Uranus and Neptune，as described in Guillot and Gautier，（2015）[58]. The interior of ice giant planets are believed to be divided into three layers，with，increasing depth（and therefore P，and T）：①a hydrogen-helium gas envelope；② ice-layers（see section 3.1.3.）； and ③ a “rock” core，possibly composed of magnesium-silicate and iron materials[58]. Neptune presents two scenarios，based on the ice to rock ratios predicted by models（see section 3.2.）. However，such models might not eventually represent the ice giant actual structures and compositions，limitations mainly exist due to the possible misassumptions of adiabatic temperature structure across interfaces[58].

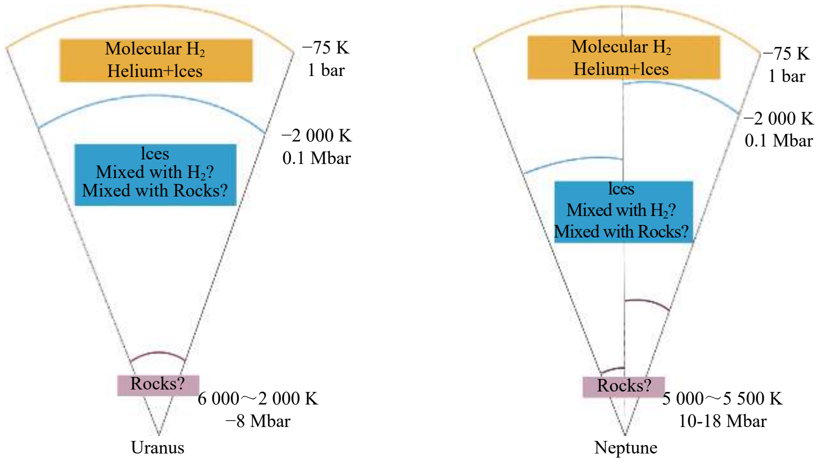

图 1　The internal structure of Uranus and Neptune，subdivided into a three-layer model：a hydrogen-helium envelope，ices，and a solid core（magnesium-silicate and iron），adapted and modified after the figure 12 of Guillot and Gautier[58]

2.1.2　The atmospheric composition

The reducing atmosphere of Neptune（and by extension Uranus）is mainly composed of ～80% hydrogen（H_2），～19% helium（He），～1.5% methane（CH_4），and traces of HD，ammonia（NH_3），ethane（C_2H_6），or acetylene（C_2H_2）[59]. Opposite to Jupiter and Saturn，the two ice giants Uranus and Neptune are characterized by a high concentration of volatiles（i.e. elements heavier than H or He）. The proportions represent ～20% for the latter，compared to ～90% for the gas giants. There are no observable solid surfaces，the atmosphere extends to great depths；it is believed that ionic oceans of aqueous ammonia may occur deep inside Uranus and Neptune[60-61]. The ice giants have strong magnetic fields，which are probably generated by dynamo currents in the electrically conductive fluids（ionic oceans of aqueous ammonia）. Whereas all present knowledge about ice giants has been deduced from remote sensing，the abundances of e.g，noble gases in the atmosphere can，however，only be measured in-situ. Therefore，most of the bulk composition of ice giants have been inferred from e.g，the abundances of their atmospheric constituents or from models[18]. In particular，models predict an enrichment in heavy elements from Jupiter to Neptune，consistent with the core accretion model[62]. Methane，and by extent the bulk C，is the only species which has been directly determined from ground-based observations（Table 1，see section 3.1.3 below）. Other remote observations were unsuccessful in determining the other volatile species such as water or ammonia[11]. Given as an example，the C/H ratio，3 times the Solar ratio at Jupiter，is supposed to be 20～30 times the Solar ratio at Uranus，and between ～30～50 times the Solar ratio at Neptune（see section 3.1.3 and Table 1 below）. Ammonia，the bulk reservoir of N in giant planets，was measured in the early 1970s by radio observations；NH_3 is found to be subsolar，unlike the gas giants，such a depletion can be explained by the presence of H_2S in the upper troposphere of the ice giants（NH_3 serves as a sink for H_2S by forming a cloud of ammonium hydrosulfide NH_4SH，see section 3.1.3 below）[11].

Besides，from the previous observations of the Galileo probe in Jupiter’s atmosphere，the enrichment in heavy noble gases at Uranus and Neptune could be explained by several hypotheses（see below）.

Table 1 Elemental abundances in the gas and ice giant planets，normalized to Solar values，based on the Table 3 from Mandt et al.[10]

Element	Jupiter	Saturn	Uranus	Neptune
He	0.8[56]	0.7 ± 0.1[68]	0.9 ± 0.2[73]	1.2 ± 0.2[77]
Ne	0.1[64]	—	—	—
O	0.4 ± 0.1[65]	（1.6 ± 0.29）× 10^{-4}[69]	—	—
C	4.3 ± 1.1[65]	9.6 ± 1.0[70]	41.5 ± 16.7[74-75]	72.1 ± 19.3[74-75]
N	4.1 ± 2.0[65]	2.8 ± 1.1[71]	—	—
S	2.9 ± 0.7[65]	12.05[72]	22.5 ± 11.3[76]	22.5 ± 11.3[78]
P	3.3 ± 0.4[66]	11.2 ± 1.3[66]	—	—
Ar	2.5 ± 0.8[67]	—	—	—
Kr	2.2 ± 0.6[67]	—	—	—
Xe	2.1 ± 0.6[67]	—	—	—

2.1.3 The enrichment in noble gases and volatiles and the cloud layers

As mentioned previously，although most of the knowledge about the bulk composition of Uranus and Neptune could be addressed by the abundances of their atmospheric constituents，e.g，C，N，S（although their uncertainties are rather large[15，18]），the noble gases could only be measured in situ by a probe，because of the absence of radio signals in the planetary atmospheres. Noble gases abundances and their isotopic ratios at Uranus and Neptune can provide insights about the origin of the building blocks which contributed to their formation[15]. Therefore，they represent valuable tools and deserve accurate measurements to understand the formation mechanisms of ice giant planets. The concentrations of noble gases in the atmosphere of Neptune have been inferred from the previous data collected by Galileo on Jupiter and are mainly based on models. The Figure 2 shows the elemental abundances in the giant planets，normalized to Solar values，based on the data presented in Table 1（after Mandt et al.[10]）. The range of variations for each element and each of the four giant planets is as well indicated. Carbon remains the only heavy element measured for all four giant planets. Its abundance increases continuously while distance with the Sun increases. Besides，an enrichment in the heavy noble gases Ar，Kr，and Xe has been identified in the atmosphere of Jupiter[63]and is therefore expected for the other gas giant and ice giant planets[64]. Such enrichments at Jupiter are，for each noble gas normalized to Solar abundances，of 0.1，2.5，2.2，and 2.1，for Ne，Ar，Kr，and Xe，respectively. Based on the measured mixing ratio of methane（the bulk C is sequestrated in CH_4），an increase of the C/H ratio of a factor of ～20～30，relative to Solar abundance，has been proposed at Uranus，and between ～30～50 at Neptune[79-80]. The C/H ratios of both Uranus and Neptune have been directly measured during the fly-by of Voyager，though with large uncertainties，and are thought to be 80 ± 20 compared to proto-Sun values[81]，but remain rather uncertain due to the assumptions made on the amount of H in non-methane volatiles[13]. Such enrichments are then expected for other condensable species.

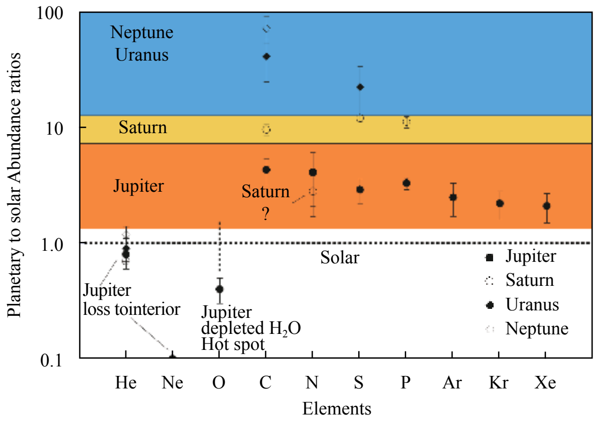

Fig. 2 Elemental abundance ratios in the atmospheres of Jupiter，Saturn，Uranus，and Neptune relative to the solar abundances，such as（*X*/*H*）observed/（*X*/*H*）Solar，where X stands for the element of interest. A solar abundance is assumed with a ratio of 1，supersolar values are indicated with ratios > 1. The color domains represent the possible range of enhancements at the considered giant planets. This figure is adapted and modified after Mousis et al.[11]

Note that both He and Ne，as observed by Galileo at Jupiter，are depleted compared to Solar：He is only 80% solar，while Ne is only 10% solar（Figure 2，Table 1）[13].

Helium condensates at pressure of 1～2 Mbars，and Ne is dissolved in liquid He；therefore，a depletion of He is characterized by a co-depletion of Ne. The condensation of He produces heat，which contributes to the internal heat of the giant planets[13]. As a consequence，the gas giant planets emit two times more heat than they actually receive from the Sun[13]. A similar trend is observed at Neptune：the ice giant emits ～2～3 times more heat than the one they receive from the Sun. However，to better understand the role of He in the heat balance of ice giants，a precise determination of the He abundance and the He/H_2 ratio is essential[13].

Some authors suggested that the noble gases could have been captured from the solar nebula at the same time when H_2 and He were captured as well[67]. Another possibility consists in evoking the "capture" of heavy noble gases，as well as CO and N_2，in clathrate hydrates. These structures might act as a "cage" and therefore trap noble gases. Such models therefore require extremely low temperatures，and an efficient clathration is only possible when assuming larger distances than the actual orbit of Jupiter[68]. Similarly，other authors evoked incorporation of noble gases into clathrates or amorphous ices[69]. These different scenarios have been reviewed in Mousis et al.[11] and are summarized hereafter in Figure 3. Assuming the gravitational instability or amorphous ice scenario，all volatiles would be identically enriched relative to Solar. The photoevaporation scenario predicts that Ne，Ar，Kr，and Xe are homogeneously adsorbed at very low temperature in the Protosolar Nebula，and a subsequent release of noble gases during the migration to the formation regions of giant planets. The clathrate scenario explains different patterns of enrichment depending on the likelihood of clathration processes，easier for e.g，Xe，CH_4 or CO_2，and more unlikely for Ar，or N_2. The last scenario，the CO snowline，explains depletion，compared to C，of species whose snowlines are beyond CO snowline. Although all these models lead to estimate the heavy noble gas abundances on the giant and ice giant planets，atmospheric entry probes directly measuring these abundances are required and remain the only option to address the noble gas composition of Neptune，and the mechanisms of enrichment predicted based on Jupiter's data scenario. In return，a detailed measurement of the noble gases would permit to appreciate the formation mechanisms of Neptune[64].

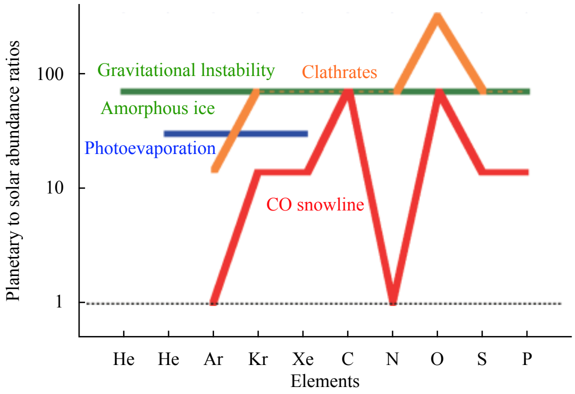

Fig. 3　The different scenarios to explain the enrichments of volatiles at the ice giants，figure adapted and modified after Mousis et al.[11]

But only direct measurements by a descent probe would be able to confirm or reject these hypotheses. One of the major goals of such an entry probe will be to measure Ar，Kr，and Xe in the well-mixed atmosphere，i.e. below the cloud layers；it is believed，due to the projected enrichments，that the droplet cloud is at ～500 bars，assuming a 50 times enrichment in condensable species compared to Solar ratio. Atreya and Wong （2004）[65] pointed out that，although at such pressures the N/H and O/H ratio would not represent the well-mixed amounts，the heavy noble gases，among others，could be determined at much reasonable depths. The so-called "Equilibrium Cloud Condensation Models"（ECCM）are used to predict the vertical structure for multi-layer clouds in the troposphere of giant planets[82-83].

Such thermochemical models，when based on the two extreme enhancement scenarios（i.e. between 30 and 50 times the Solar ratios），predict the altitude at which the different cloud layers could be located. From top to depth，models suggest the presence of a methane-ice cloud layer at ～1.5 bar；Voyager 2 effectively "observed" a cloud layer located at ～1 bar level. It is followed by an ammonia-ice cloud at ～10～13 bars. Based on such ECCM models，below this cloud，NH_3 is expected to be well-mixed and close to Solar value，but certainly not depleted by factors of ～100 as previously observed[13]. As previously mentioned，such depletion in NH_3 can be explained by the presence of a H_2S cloud in the upper troposphere of the ice giant，due to the fact that

NH_3 is a sink for H_2S，by forming a cloud of NH_4SH. The water-ice cloud might be encountered at pressure of ～53 bars. Assuming an enrichment of 50 times the Solar ratio，the water-ammonia aqueous solution cloud could be encountered at depths of ～88 bars（Figure 4）.

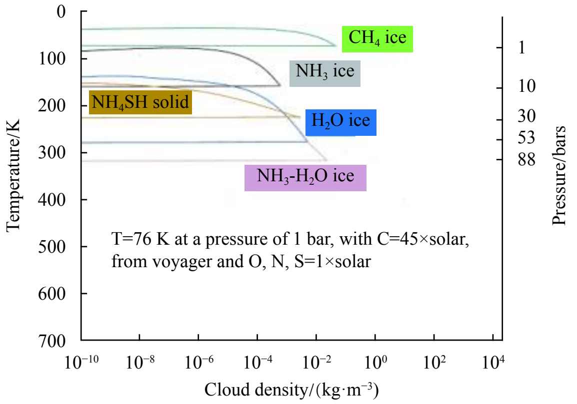

Fig. 4 Structure of the cloud layers in the ice giants based on Equilibrium Cloud Condensation Models（ECCM），based on Weidenschilling and Lewis[83]. The assumptions are the following：a temperature of 76 K at a pressure of 1 bar，an enrichment in C of 45 times the Solar value（Voyager data[84]），and O，N，and S are considered to be equal to Solar value. This figure is adapted and modified after the Figure 5 of Atreya et al.[13]

Going deeper within Neptune's interior，some models even suggest the presence of ionic/superionic oceans of aqueous ammonia at kilobar[61] to megabars pressures[65]. Assuming the existence of such an "ocean"，it would imply that water and ammonia would dissolve in it，therefore causing the observed "depletion" of those mentioned species above that ocean. As a matter of fact，considering this hypothesis as true would have significant effect regarding the design of the probe mission，as measurements to only ～10-20 bars，rather than kilobar levels，would be necessary[86]. These authors mentioned that，consequently，all heavy elements，such as，among others，Ar，Kr，Xe，but as well C，or S can all be measured at pressures of less than 20 bars. Measurements of such elements are necessary for our comprehension of Neptune's atmospheric dynamics and will help to improve and constrain the formation models.

2.2 The isotopic ratios and the D/H ratio：insight on the interior of the ice giants

Little is known about the volatile isotopic composition at the ice giant planets. On one hand，as discussed earlier，the measurements of noble gas ratios，in particular，would provide a significant insight about the possible atmospheric evolution of Uranus and Neptune. It would be especially interesting to verify if their isotopic composition is，like Jupiter，similar to that of the Sun（Table 2），or if it has been modified by some post-formation processes[13]. Indeed，loss of volatiles since the formation of gas giant planets is unlikely，given their large mass and strong magnetic fields. Therefore，their composition might represent the initial conditions of the primordial Solar Nebula[13]. On the other hand，the D/H ratios might give insight about the ice to rock ratio at Uranus and Neptune. The D/H ratios in the two planets have been derived from ground-based measurements at Herschel-PACS[86]，and are rather close to the protosolar value；the D/H ratio at Neptune is in the range of（4.1 ± 0.4）× 10^{-5}，which is as well really close to the one expected for Uranus，i.e.（4.4 ± 0.4）× 10^{-5}（Table 2）. We report as well in Table 2 the elemental isotopic ratios which bring information on the formation and evolution processes of our Solar System：$^{13}C/^{12}C$，$^{15}N/^{14}N$，$^{3}He/^{4}He$，$^{20}Ne/^{22}Ne$，$^{38}Ar/^{36}Ar$，as well as those of Xe. The elemental isotopic composition at Jupiter measured by the Galileo probe are in Solar proportions（c.f. Table 2，and e.g，Atreya et al.[13]）. A precise determination of the D/H is necessary in order to make constraints on the protosolar ratio，which still remains uncertain. Indeed，this D/H ratio is not yet measured but derives directly from the $^{3}He/^{4}He$ ratio measured in the Solar wind，to which the inferred primordial $^{3}He/^{4}He$ ratio is subtracted -this one is extrapolated from the $^{3}He/^{4}He$ ratio measured in meteorites and Jupiter by Galileo[11]. Therefore，precise $^{3}He/^{4}He$ measurements at Neptune would serve as benchmarks regarding the protosolar D/H ratio. In addition，a precise measurement of D/H would allow models to have a better view of the interior of Neptune，and more precisely the ice-mass fraction. Based on the D/H of（4.1 ± 0.4）× 10^{-5} at Neptune would imply an ice-mass fraction of ～14%～32%，therefore it is an argument in favor of Neptune's interior（and by extension Uranus's interior）being more rocky than icy[13]，in contradiction with previous thoughts（25% rock-dominated，vs. 60%～70% ice-dominated[87]）. In addition，the D/H ratio signature might constrain the environment on the Solar Nebula from where the planet has formed. As explained in Yang et al.[88]，the expected

D/H ratios of water in the inner Solar Nebula would reach $\sim 2 \times 10^{-5}$ whereas it is expected to be lower for a formation occurring in the outer disk，due to slower rates of isotopic exchanges. The D/H ratio at Uranus and Neptune has as well implications on bulk oxygen abundance. In another study from Ali-Did et al.[89]，the measured chemical composition for Neptune（D/H and C/H）was used to predict the oxygen abundance.

Table 2　Elemental isotopic ratios in the Sun Jupiter，Saturn，Uranus，and Neptune，based on the Table 3 from Atreya et al.[13]

Elements	Sun[90]	Jupiter[91]	Saturn[91]	Uranus[88]	Neptune[86]
D/H	$(2.0 \pm 0.4) \times 10^{-5}$	$(2.6 \pm 0.7) \times 10^{-5}$[92]	$(1.70^{+0.75}_{-0.45}) \times 10^{-5}$[93]	$(4.4 \pm 0.4) \times 10^{-5}$	$(4.1 \pm 0.4) \times 10^{-5}$
$^{12}C/^{13}C$	0.0112	0.0108 ± 0.0005	0.0109 ± 0.001	—	—
$^{14}N/^{15}N$	$(2.27 \pm 0.08) \times 10^{-3}$	$(2.30 \pm 0.03) \times 10^{-3}$	$<2.0 \times 10^{-3}$	—	—
$^{36}Ar/^{38}Ar$	5.50 ± 0.01	5.60 ± 0.25	—	—	—
$^{136}Xe/Xe$	0.0795	0.076 ± 0.009	—	—	—
$^{134}Xe/Xe$	0.0979	0.091 ± 0.007	—	—	—
$^{132}Xe/Xe$	0.2651	0.290 ± 0.020	—	—	—
$^{131}Xe/Xe$	0.2169	0.203 ± 0.018	—	—	—
$^{130}Xe/Xe$	0.0438	0.038 ± 0.005	—	—	—
$^{129}Xe/Xe$	0.2725	0.285 ± 0.021	—	—	—
$^{128}Xe/Xe$	0.0220	0.018 ± 0.002	—		
$^{20}Ne/^{22}Ne$	13.6	13 ± 2	—	—	—
$^{3}He/^{4}He$	1.66×10^{-4}	$(1.66 \pm 0.05) \times 10^{-4}$	—	—	—

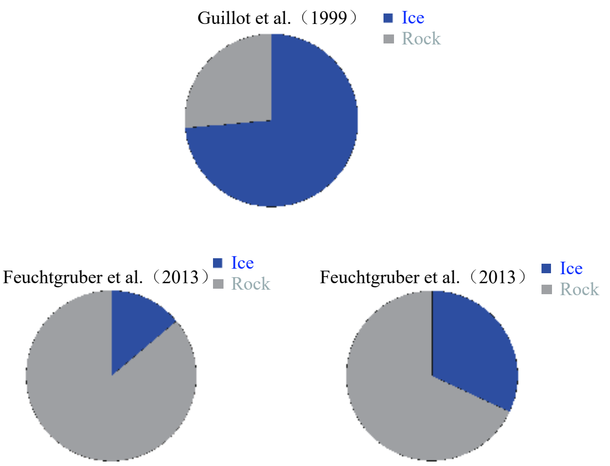

Fig. 5　The different scenarios for different D/H ratios at Neptune[86-87]，and the partition between ice and rock composition of Neptune's interior. From the inferred D/H ratio of Guillot et al.[87]，Neptune's core is 25% rock and between 60%～70% ice-dominated，whereas models presented in Feuchtgruber et al.[86] would imply a partition $F = 0.14 \sim 0.32$，and therefore 68%～86% of heavy components are made of rock，whereas 14%～32% consist of ice

3　The proposed mission profile

3.1　The payloads onboard an interstellar mission

As mentioned earlier in this review，the ice giants represent a challenge in our comprehension of the evolution of the Solar System. Uranus and Neptune are the least studied planets of our Solar System，though most of the newly detected exoplanets are Neptune-sized planets[13]. Having this in mind，the exploration of the ice giants has been prioritized for future interstellar missions，and the feasibility of such entry probes discussed in various space agencies decadal surveys. A detailed description of the scientific goals and instruments onboard an atmospheric entry probe to Neptune is presented in e.g，Bienstock et al，Mousis et al.，Atreya et al. or Vorburger et al.（2020）[64，11，13，18]. Bienstock et al.[64] listed six major scientific objectives，including① the measurements of elemental ratios，relative to hydrogen，and the key isotopic ratios，e.g，D/H，$^{13}C/^{12}C$，$^{15}N/^{14}N$，$^{3}He/^{4}He$，$^{20}Ne/^{22}Ne$，$^{38}Ar/^{36}Ar$，as well as those of Xe；② the planetary processes，including the chemistry of the atmosphere，the cloud layers，or the winds；③ the investigation of Neptune's captured moon Triton，with an emphasis on its atmospheric composition，and the geological processes；Triton is，with Io，Enceladus，and of course the Earth，one of the four bodies of the Solar System with -observed- eruptions of materials；④ the rings：their origin，evolution，and structure；⑤ the "magnetospheric and plasma processes"；and ⑥ the study of Neptune's icy satellites（composition，geology， etc.）. Several mission concepts to Neptune and the Neptunian system have been proposed through the past decades. As an example，the NASA’s mission *ARGO*，which had an initial launch window between 2015—2020，would have aimed to reach the Neptunian system within ～8～11 years，with flybys of Jupiter and Saturn，followed by a 6-month science at Neptune and Triton around 2029. The general trajectories would have been the same as those of Voyager，previously. Then the mission would have later focused on the KBOs around 2033[94].

Concerning now the payload instruments onboard the entry probe，a comprehensive review has been made by e.g，Bienstock et al，Hansen et al，or Mousis et al.[65，94，11]. The proposed atmospheric entry probe to Neptune could be equipped，among others，with a Gas Chromatograph Mass Spectrometer（GCMS）for the measurement of noble gas abundances，isotopic ratios，as well as D/H ratio[64]，an atmospheric structure instrument，a helium abundance detector[11，64]，a near infrared-spectrometer. The latter was not existing at the time of Voyager and would help provide the distribution of CH_4，CO，and CO_2 ices，in order to address e.g，the volatile transport on Triton and the KBOs[94]. As the winds in Neptune are by far the strongest in the whole Solar System，Bienstock et al. and Mousis et al.[64，11] both suggest having a Doppler Wind Experiment（DWE）onboard the probe. Similarly to the Huygens Atmospheric Structure Instrument onboard the Huygens probe on Cassini mission to investigate Titan’s atmospheric structure and electrical properties[95-96]，Mousis et al.[11] suggest equipping the probe with an Atmospheric Structure Instrument（hereafter called ASI）. It would consist in a multi-sensor package，including e.g，accelerometers，pressure profile instrument，temperature sensors，Permittivity Wave & Altimetry，which goals are to measure the physical properties of the atmosphere of Neptune，including temperature（T），pressure（P），density，or conductivity. During the probe’s descent，the ASI will determine，among others，the P and T profiles. On the orbiter，Bienstock et al.[64] opted for e.g，a magnetometer; indeed，Voyager 2 discovered that Neptune’s magnetic field is 27 times more powerful than on Earth and in addition changes periodically when interacting with the solar wind. Additionally，the orbiter could be equipped with high-resolution UV and IR spectrometers，a high-resolution camera，a mid- and far IR spectrometer，a laser altimeter，a microwave radiometer，or a penetrating radar.

3.2 The mass spectrometers for the detection of volatiles and noble gases：a review

Being interested in the noble gas and the volatile abundances of the ice giants，we will discuss here into more details the atmospheric probing mass spectrometer onboard a potential entry probe to Uranus and/or to Neptune. Mass spectrometers are instruments which can provide a unique set of data，being sensitive and quantitative，about the chemical composition of the atmosphere，the volatile isotopic ratios，as well as the elemental and molecular abundances of species of interest. Willing to have the most accurate data as possible，such an ideal mass spectrometer would，on one hand，possess a high sensitivity，dynamic range，resolution，and accuracy，and on the other hand would have a minimum weight，with reasonable dimensions，and would be energy-efficient. Facing the extreme environment of Uranus or Neptune，the instruments onboard the entry probe must resist to high variations of temperatures and pressures，as well as high G-forces[18]. Today’s newly generation of mass spectrometers developed for future space missions can reach mass resolutions $m/\Delta m$ ranging from $9\,000 < m/\Delta m < 100\,000$（e.g，Vorburger et al.[18]，and references therein）.

Table 3 hereafter summarizes the different payloads，mass spectrometers and other complementary instruments，as well as their characteristics，such as mass resolution，and their possible use，advantages and counterparts，onboard an interstellar mission for the investigation of volatiles at the ice giants. Note that a detailed review about mass spectrometry in space is available in Vorburger et al.[18]. The time-of-flight（TOF）mass spectrometers and the compact Orbitrap represent the two high-resolution mass spectrometers，particularly suitable for accurate measurements，and capable of separation of adjacent masses as well as resolving isobaric interferences. However，an ion trap or a quadrupole represents a really good compromise between high mass resolution and compact design. Given the long travel period of an interstellar mission to the Outer Solar System，payloads with low power consumption and weight are much more favorable. We give here five examples of mass spectrometers which have equipped five different space missions. It is not here our intention to discuss the results in detail，but instead to give a short overview of the past missions equipped with different mass spectrometers.

Table 3　Comparison of the different types of instruments used for noble gas and volatile measurements

Mass Spectrometers	Mass resolution$m/\Delta m$	Characteristics
Time-of-flight-mass spectrometer（TOF）	~1 000* to 30 000[97] 350 000[98]	- Can achieve really high mass resolution- High mass range
Magnetic sector mass spectrometer		
Orbitrap	100 000 with m/z = 400$m/\Delta m \sim m^{-1/2}$	- Achieve really high mass resolution- Compact design
Quadrupole and ion trap	<1 000	- Sufficient resolution to separate adjacent mass lines- Cannot resolve isobaric interferences
Other complementary instruments		
Gas Chromatograph Mass Spectrometer（GCMS）		- Powerful analysis method- Equipped Viking，Cassini-Huygens，MSL on Curiosity，etc.
Tunable Laser spectrometer（TLS）		- Isotopic ratios of selected molecules- Ultra-high spectral resolution- High accuracies（few % for species，few ‰ for isotopes）
Helium Abundance Detector（HAD）		- Complementary to mass spectrometer- Very compact design- Energy-efficient

*ex. of RTOF onboard ROSINA，which has a $m/\Delta m$ = 5 000

1）The example of the Galileo Probe Mass Spectrometer（GPMS）which investigated Jupiter in 1995，and revealed the composition of the Jovian atmosphere，can be viewed as an analog for the exploration of the ice giants. Among others，the He mole fraction measured in the Jovian troposphere by the HAD was found to be 0.1359 ± 0.0027，in really good agreement with the GPMS[54-55]（see section 1.2. for more details）.

2）Another example is NASA's Viking Project，which was the first U.S. mission to land a spacecraft safely on the surface of Mars. The very first geochemical exploration of Mars has been conducted by the two Viking landers. The Martian atmospheric gas composition has been performed by mass spectrometers embarked aboard the Viking landers[47-48]. Measurements of the CO_2，H_2O，N_2，O_2，the noble gas abundances，as well as ratios of the isotopes of hydrogen，carbon，oxygen，nitrogen，argon，krypton，and xenon，have been acquired. A more detailed review about Martian atmospheric measurements can be found in Ott et al.[99]. Similarly to the GPMS，the instrument onboard the Viking probe has a high sensitivity，a high mass range，and a high resolution，to resolve the minor components of the Martian atmosphere. Gas scrubbers have been used to reduce the partial pressures of species such as CO and CO_2，too abundant in the Martian atmosphere，and which would compromise the accuracy of measurements. After a combination of 15 measurement cycles，the signal has been increased by a factor of 5.3. The very first noble gas isotopic ratios were obtained，however some with large uncertainties：^{36}Ar/^{38}Ar（5.5 ± 1.5），^{40}Ar/^{36}Ar（3 000 ± 500），or ^{129}Xe/^{132}Xe（2.5^{+2}_{-1}）[48].

3）Still related to the exploration of the Martian atmosphere，the Mars Science Laboratory（MSL）mission onboard the Curiosity rover was equipped with a suite of scientific instruments with，among them，the Sample Analysis at Mars（SAM）. The SAM suite onboard the Curiosity rover included a quadrupole mass spectrometer（QMS），a tunable laser spectrometer（TLS）and a 6-column gas chromatograph（GC）system[49-50]. Precise noble gas isotopic ratios were measured，including ^{36}Ar/^{38}Ar（4.2 ± 0.1），^{40}Ar/^{36}Ar（1 900 ± 300），^{84}Kr/^{86}Kr（3.269 ± 0.074），or ^{129}Xe/^{132}Xe（2.524 ± 0.002）[49-50].

4）The Cassini-Huygens mission was equipped with a Gas Chromatograph Mass Spectrometer（GCMS）onboard the Huygens entry probe to Titan. The combination of Gas Chromatography（GC）and mass spectrometry offers the advantage of separating the mass spectra；the GC separates the species which are later identified by the mass spectrometer. The main components of a GCMS consist in ① a mass spectrometer，equipped with five ion sources；② a gas sampling system，with direct atmospheric sampling and enrichments of trace species or noble gases；③ the GC；and ④ sample transfer system to the mass spectrometer[57]. Argon-36 as well as ^{40}Ar were detected，the other primordial noble gases being below 10^{-8} mixing ratio；isotopic ratios of $^{12}C/^{13}C$，$^{14}N/^{15}N$ or D/H were determined. In addition，altitude profiles were acquired for key volatiles such as CO2，ethane，or cyanogen.

5）More recently，ROSINA（Rosetta Orbiter Spectrometer for Ion and Neutral Analysis）onboard Rosetta mission carried out measurements of the elemental and isotopic compositions of the comet 67P Churyumov-Gerasimenko（67P/C-G），as described in e.g，Luspay-Kuti et al，Le Roy et al，Mall et al，or Marty et al.[100-103]. The ROSINA is composed of two mass spectrometers，a Double Focusing Mass Spectrometer（DFMS），and a Reflectron Time-of-Flight mass spectrometer（RTOF）. The measurements began since the spacecraft arrived at its destination in August 2014 and were ongoing for two more years. The DFMS has a high mass resolution（the mass divided by the change in mass，here 3 000 at 1% peak height，at a mass over charge ratio of $m/z = 28$），with a $m/\Delta m = 9，000$ at 50% peak height，covering a mass range of $12 < m/z < 150$. Heterogeneities in the composition of the coma（gases emitted from comet 67P/C-G by sublimation of ice）as well as diurnal variations in the compositions have been observed. Nitrogen- and sulfur-bearing species，as well as oxygenated compounds were detected[101]. With such a high mass resolution，Xe elemental and isotopic measurements（at $m/z = 129$，131，132，134，and 136）have been carried out essentially without any interference at similar mass[103]. Calibrations have been performed in-flight，using reference gases. Reference gases are required in order to calibrate the measurements，i.e. to have access to the detector gain or mass scale determination，and especially after a long journey into space，which would be the case for an interstellar probe visiting Uranus or Neptune. On the other hand，RTOF had a slightly lower mass resolution（i.e. $m/\Delta m = 5\,000$）but was able to cover a greater mass range（$1 < m/z < 1\,000$[104]）. The combination of both DFMS and RTOF lead to the detection of more than 100 different species，including noble gases，or organic compounds[18]. Ptolemy onboard the Rosetta mission measured the stable isotopic ratios of light elements：$^2H/^1H$（via H_2O），$^{13}C/^{12}C$，$^{15}N/^{14}N$，$^{17}O/^{16}O$，and $^{18}O/^{16}O$（see section 3.3 hereafter）.

3.3 The noble gas and volatile measurements

Here we present，based on the key questions raised in this review and/or in associated volatile-noble gas-related research，possible scenarios for an entry probe to one of the ice giants. We decide to focus here on Neptune. The choice of Neptune over Uranus（or vice-versa）as a target for exploration is still under debate. Science at Neptune would as well give access to the KBOs. However，none of the two ice giants has our preference here. Two recently published reviews from Jarmak et al. and Sayanagi et al.[12，17]，however focused on a mission concept to Uranus. The following species and isotopic ratios（non-exhaustive）are of interest when considering the processes at the origin of the ice giant formation：CH_4，NH_3，H_2，HD，H_2O，CO，CO_2，$^{13}C/^{12}C$，$^{15}N/^{14}N$，$^{16}O/^{17}O$，$^{18}O/^{16}O$，He and the $^3He/^4He$ ratio，Ne and the $^{20}Ne/^{22}Ne$ ratio，Ar and the $^{38}Ar/^{36}Ar$ ratio，and heavy noble gases，the isotopes of Kr and as well those of Xe[18]. A precise description of a descent probe has been discussed in Vorburger et al.[18]，with，similarly，an emphasis on the ice giants' chemical composition，volatiles，and noble gas abundances. Ideally，the scientific instruments onboard a probe require the highest sensitivities，accuracies，and mass-resolution in order to acquire as much precise data as possible during the descent phase. However，TOF-MS，despite lower mass resolutions，could cover great mass range，therefore suitable for volatile measurements. A special feature of such a probe is to maintain a stable internal pressure while external pressure varies by several orders of magnitude during descent. Based on the heritage of Galileo at Jupiter in 1995，the mission lasted for about ～75 minutes before termination，the probe reaching a pressure of ～20～25 bars and covered about 0.22% of Jupiter's radius. In the harsh environment of Neptune，the goal of such an entry probe would be to reach a minimal pressure of ～0.1 bar，which represents the interface between the stratosphere and the troposphere，the latter being considered as homogeneously mixed. It is expected that the probe will pass through the stratosphere and reach a depth of at least ～10～20 bars，in order to obtain the complete composition and study the dynamics of the Neptunian atmosphere[84]. Vorburger et al.[18] suggest equipping the descent probe with a system subdivided into ① a gas separation and enrichment system；② a reference gas system；③ a mass spectrometer； and ④ a tunable laser spectrometer（TLS）. We will here briefly describe each of those sub-systems. More details can be found in Vorburger et al.[18].

1）As part of the volatiles in the stratosphere/troposphere will be in condensed phase，the gas inlet system should be heated properly in order to systematically volatilize all incoming condensed particles. The gas separation and enrichment system include cryotraps and getters（non-evaporable getters）to collect and purify e.g，noble gases，and，similarly，to remove all other components which could otherwise compromise the accuracy of the measurement. It is estimated by Vorburger et al.[18]that cryotraps and the whole procedure will contribute to enrich the noble gases（and especially the heavy noble gases）by factors of ～5 000.

2）The reference gas system contains reservoirs filled with different gases of known isotopic and chemical compositions.

3）The mass spectrometer proposed by Vorburger et al.[18] is a TOF mass spectrometer with a low resolution of ～1 000，which is sufficient for the separation of ^{3}He and HD（$m/\Delta m$ ～500）. However，an ion trap is also suitable for the task while high RF frequency is applied. In addition to the only TOF-MS for the measurement of He，a HAD can be added，keeping in mind that a precise determination of the He/H_2 abundances is necessary，as for the Galileo mission at Jupiter[54-55]. Precision of the He mole fraction by the HAD is still better than with a mass spectrometer.

4）The TLS will directly sample the gas and measure isotopic ratios of interest，such as e.g，D/H，$^{13}C/^{12}C$，$^{15}N/^{14}N$，$^{16}O/^{17}O$，$^{18}O/^{16}O$，etc.

The suite of scientific instruments onboard an interstellar Chinese mission to the outer Solar System have not yet been decided. However，the possibility of embarking an ion trap mass spectrometer，（ITMS）is a good compromise. Ion traps are compact，they are among the smallest flight mass spectrometers，energy-efficient，and have sufficient mass resolution to separate adjacent mass lines. They are a compromise to QMS，capable of in-situ characterization of planetary environments（see discussion below）. Some new developments lead to increase mass resolution up to $m/\Delta m$ =12，000 at 40 Da[105]. Xenon isotopic measurements have already been carried out using the Jet Propulsion Laboratory Quadrupole Ion Trap（JPL-QIT）[106]. The main objective was to determine the instrument viability and stability under specific conditions（P，T），in the context of future planetary science missions targeting accurate isotopic measurements of heavy noble gases and light elements[107]. Isotopic composition of commercial standards has been measured；the obtained results point to good reproducibility and accuracy（better than 0.07%）.

We give now more information about implementation of ITMS in latest space missions. Ion traps have already been implemented in two major space missions[107]，and are as well used for on-orbit measurements of the atmospheric constituents aboard the International Space Station（ISS）[108]（Table 4）.

Table 4 List of ion trap mass spectrometers（ITMS）in space or under development，mass range and resolution（$m/\Delta m$）

Space missions	Year	Name	Characteristics
Team			
Open University	2004—2015	Rosetta-Ptolemy	Mass range：12～150 Da$m/\Delta m$ = 150
NASA-ESA	2018	ExoMars-MOMA	Mass range：50～1 000 Da$m/\Delta m$ = 50～500
ISS mission			
JPL	2010—2012	VCAM	Mass range：15～100 Da$m/\Delta m$ = 220
JPL	2019	SAM	Mass range：10～300 Da$m/\Delta m$ = 800
Further developments			
NASA	—	LITMS	Mass range：20～2 000 Da
JPL	—	MARINE	Mass range：10～320 Da$m/\Delta m$ = 750$m/\Delta m$ = 4 000 @ 10～80 Da

（1）The first onboard space borne ITMS was Ptolemy（Open University，2004—2015）for the Rosetta mission. It consisted of a GCMS，centered on a 3-D ion trap[107]. It measured the stable isotopic ratios of light elements：$^{2}H/^{1}H$（via H_2O），$^{13}C/^{12}C$，$^{15}N/^{14}N$，$^{17}O/^{16}O$，and $^{18}O/^{16}O$. Some of its technical characteristics are summarized in Table 4 hereafter.

（2）The Mars Organic Molecule Analyser（MOMA），a 2-D ITMS，onboard the ExoMars mission（2018，NASA-ESA）[107].

（3）Aboard the ISS，trace gas and major atmospheric constituents have been measured by e.g，the Vehicle Cabin Atmosphere Monitor（VCAM）；volatile trace gas components were measured at ppm to ppb levels，as well as the major atmospheric constituents（N_2，O_2，Ar，CO_2）[108].

（4）Similarly，NASA’s Spacecraft Atmosphere Monitor（SAM）aboard the ISS aimed at measuring the atmosphere quality aboard the space station. SAM is one of the smallest autonomous GCMS instruments ever built，and detects major components found in air，such as O_2，CO_2，N_2，and CH_4，as well as monitors humidity levels in real-time（NASA’s website）.

Efforts are currently undertaken to develop ITMS for future space mission applications. Linear Ion Trap Mass Spectrometers（LITMS），a direct heritage of the MOMA ITMS，will be capable of the detection of organic and inorganic species over expanded mass range（Table 4），positive and negative ions，among others[107].

The Jet Propulsion Laboratory（JPL）develops the Mass Analyser for Real-time Investigation of Neutrals at Europa（MARINE），a 3-D ITMS characterized by low mass，low power，and ultra-high sensitivity and precision measurements；its main objective is the measurement of the abundance of neutral gases（H_2O，O_2，CO_2，SO_2）in the exosphere of Europa[107].

Quadrupole mass spectrometers have been implemented in series of space missions from NASA or from India（see section 3.2 and Table 5）：e.g，Pioneer mission to Venus[109]，GPMS onboard Galileo to Jupiter[52]，GCMS onboard Cassini-Huygens to Titan[56]，SAM onboard Curiosity to Mars[49-50]，NGIMS onboard MAVEN to Mars，or MENCA onboard MOM to Mars.

-If we now compare the performances of both ITMS and QMS（Table 5），we can draw the following conclusions：Both have similar resolution（～1 amu）

-QMS have higher dynamic range，2-4 orders of magnitude higher（Table 5）

Table 5　Comparison of performances between ITMS and QMS

payload	Name	Space missions（Team）	Mass/kg	Power/W	Mass range/Da	Dynamic range	Sensitivity/（ct·s^{-1}·Torr^{-1}）
ITMS[108-109]	Rosetta-Ptolemy	67P/C-G	4.5（MS：0.5）	10（MS：1）	12～150	—	1×10^{10}
	ExoMars-MOMA	Mars	9.3	70	50～1 000	—	—
	VCAM	ISS	30.3（MS and pumps：5.4）	105（MS：42）	15～100	—	2×10^{12}
	SAM	ISS	9.55	45	10～300	—	2×10^{13}
	LITMS	NASA			20～2 000	—	—
	MARINE	JPL	7.3	14	10～320	1×10^{6}	1×10^{15}
QMS	ONMS[108]	Pioneer（Venus）	3.8	12	1～46	—	2×10^{13}
	NMS[109]	Nozomi/Planet-B（Mars）	2.8	7.4	1～60	—	3×10^{12}
	GPMS[52]	Galileo（Jupiter）	13.2（with pump）	13（+12）	2～150	1×10^{8}	—
	GCMS[55]	Cassini-Huygens（Titan）	17.3（with pump）	110	2～141	1×10^{8}	1×10^{14}
	SAM[49-50]	Curiosity（Mars）	40（all）	175	2～535	—	2×10^{14}
	INMS[110]	Cassini Orbiter（Saturn and satellites）	10.3	23.3	1～99	1×10^{8}	2×10^{14}
	NMS[111]	LADEE（Moon）	11.3	34.4	2～150	1×10^{8}	6×10^{14}
	NGIMS[112]	MAVEN（Mars）	12	36	2～150	1×10^{8}	6×10^{14}
	MENCA[113]	MOM（Mars）	3.56	29	1～300	1×10^{10}	—

-QMS have as well better accuracy for isotope measurements

-However，ITMS are more compact（therefore lighter）and consume less power

It is reasonable to assume that ITMS will be preferred in future space exploration because they can reach a much higher mass limit at a reasonable power consumption[114]，which is very attractive in identifying large molecules that might indicate prebiotic chemistry and biosignatures.

3.4 An entry probe mission scenario

After about 13 years journey，the proposed mission is expected to reach Neptune，time when the entry probe could be released in the equatorial zone of the planet，using the fact that，at such latitude，Neptune rotates faster，which can reduce the entry speed of the probe，thus expected to be between ～22～26 km/s. In addition，atmospheric circulation mechanisms must be understood and should be considered as they can provide crucial information as for the choice of a suitable entry for a future in situ probe mission to Neptune. They could be achieved by remote sensing（observation of the reflected sunlight in the ultraviolet，observation of thermal emission in the mid/far-infrared，or observations of thermospheric emission from H_2 and H_3^+）. Although noble gas abundances and their isotopic ratios could be measured at any latitudes，the chemical enrichments in volatiles species and precise determination of other isotopic ratios of interest require a sampling in a representative region of the ice giant. Therefore，Fletcher et al.[115] suggest avoiding the low latitude-detected meteorological features，aiming preferentially an entry at an off-equatorial region of the planet.

Upon arrival，the descent will be carried out with the assistance of a parachute，similarly to the Galileo mission，and the very first set of data will be collected at different depths，and returned to Earth via a relay station：composition，structure，and dynamics of the atmosphere. A detailed review about a possible NASA/ESA collaboration mission profile is available in Mousis et al.；Vorburger et al.[11，18] described as well a scenario in which a descent probe collects data about atmospheric compounds at one the ice giants；the operations are then divided in several phases，during which atmosphere is analyzed during，first，during the probe free-fall（with，here，a special an emphasis on the H_2/He ratio，one of the most valuable data）. After the parachute deploys，the integration time becomes greater and therefore measurements are more suitable for a high vertical resolution mode. Reaching the top cloud layers at P ～1 bar（CH_4 ices），instruments would operate at the maximum of their sensitivities，in order to increase detection of the rarest atmospheric components，as well as noble gases and their isotopic ratios. Finally，the loss of communications between the entry probe and the orbiting spacecraft are expected to happen ～1 hour after the first measurements.

The choice of Neptune over Uranus（or vice-versa）as a target for exploration is still a debate topic. The final decision might turn into the favor of Neptune，given the argument that science at Neptune would as well give access to the KBOs；among them，Neptune’s moon Triton，the seventh largest moon in the Solar System，is apparently made of the same materials as Pluto（i.e. crystalline and amorphous ices of CH_4，N_2，CO），and is therefore believed to have been captured by the planet. Triton’s orbit is retrograde，and it orbits Neptune in the opposite direction of its parent planet. A simultaneous investigation of Neptune and Triton，via volatiles transport on this moon and other KBOs，might reinforce the choice of Neptune as a candidate for exploration. As mentioned in a 2009 Planetary Science Decadal Survey[116]：“Neptune and its captured moon Triton are unexplored with modern spacecraft instrumentation. Observations of these objects are urgently needed to address planet formation and the evolution of ice giant planets，icy satellites，Kuiper Belt Objects，and the solar system itself”.

4 Conclusions

Our understanding of Neptune，and more particularly of the ice giants，is so far restricted to the only data which have been collected by the flyby of the last planet of our Solar System in 1989 by the Voyager 2 probe. The increasing number of detected ice giant-like exoplanets consequently reinforces the limits of our knowledge about ice giants，their formation，evolution，and atmospheric dynamics. Indeed，unanswered questions remain present，mostly about the composition of the Neptunian atmosphere，especially the noble gas isotopic composition，and key isotopic ratios，such as the D/H ratio，etc. The latter is of particular importance since a precise determination of D/H might better constrain the existing models，as well as our comprehension of the internal structure of the planet. In that context，the feasibility of initiating a deep space and interstellar exploration in the outer Solar System represents a unique opportunity to deliver an atmospheric entry probe to Neptune，and thus to tackle all these questions. The designed probe，equipped with，among others，a TOF/gas chromatograph mass spectrometer，a tunable laser spectrometer，an atmospheric structure instrument，a helium abundance detector，or a near infrared-spectrometer，would concentrate on collecting data which are crucial in our understanding of atmospheric composition and dynamics of ice giants. Ion trap mass spectrometers represent a good compromise between high mass resolution and compact design；they are candidates to be part of a suite of other scientific instruments onboard a future interstellar Chinese mission to the outer Solar System.

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海王星探测：稀有气体和挥发分分析

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摘　要：地外天体的探测目标主要是太阳系中离地球较近的行星和天体，例如水星、金星、月球、火星、木星、小行星67P和近地小行星25143糸川。目前，人类还没有明确提出对太阳系中两大冰巨行星天王星和海王星的探测计划。人类探测仍停留在“旅行者2号”探测器分别于1986年1月和1989年8月飞掠天王星和海王星时传回的数据。在过去的几十年间，越来越多类属冰巨行星的系外行星被发现，而且冰巨行星比类似木星和土星的气态巨行星数量更多，加深对冰巨行星的了解势在必行。其中，行星大气组成，特别是稀有气体和挥发分的分布尤为重要。详细回顾了对太阳系中各天体挥发分的探测及结果；总结了天王星和海王星的稀有气体和挥发分的浓度、分布和演化过程；讨论了探索冰巨行星的星际探测任务的流程、可行性以及探测器载荷选择。提出了利用离子阱质谱仪作为中国外太阳系探测任务中探索冰巨行星科学载荷的可行性。

关键词：冰巨行星；海王星；挥发分；稀有气体；同位素比值；质谱仪；进入探测器

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(上接第566页)

One-Dimentional Numerical MHD Simulation of the Shock Events in the Outer Heliosphere

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Abstract：Voyager 1 occasionally detected the sudden jumps of the interstellar magnetic field strength since its heliopause crossing in August，2012. These events are believed to be the interstellar shocks，and associated with the product of the interaction between the large-scale solar wind structures and the heliopause. In this paper, this possibility is examined by means of a two-fluid magnetohydrodynamics（MHD）simulation consisting of the solar wind plasma and the interstellar neutrals. Three different solar wind observations from OMNI，STEREO A and B at a heliocentric distance of 1 au during the year 2010－2017 are used as input to the simulation，and the evolution of solar wind in the outer heliosphere is investigated after the charge-exchange between solar wind and interstellar neutrals.The numerical results are compared with the observation from the two Voyagers，showing that the shock events in the interstellar medium observed by Voyager 1 have direct linkage with the pressure pulses in the inner heliosheath detected by Voyager 2.

Keywords：outer heliosphere；shock events；solar wind；numerical MHD simulation

Highlights：

- It is the first time to use the solar wind data from OMNI，the spacecraft STEREO A and B orbiting the Sun conjunctly to study the evolution and propagation of the solar wind in the outer heliosphere.
- The numerical simulations have been carried out to verify the possible correspondence between the large-scale pressure pulse structure in the heliosphere and the shock event in interstellar space.

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