Noble Gases

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Without Abstract

Definition

The noble gases occupy Group 0 (column 18) of the periodic table, comprised of the following elements: helium, neon, argon, krypton, xenon, and radon (not discussed here). They are colorless, odorless, monatomic, and unreactive gases at standard temperature and pressure.

Introduction: History and Uses

Helium **:** The spectral signature of helium was first observed from the Sun by Janssen and Lockyer in 1868, with Lockyer credited for proposing this as a new element. Helium was first isolated and identified in clevite, a uranium-rich mineral, by both Cleve and Langlet and independently by Ramsay in 1895. ^{4}He is formed by alpha particles during the decay of natural uranium and thorium in the crust, with commercial sources provided in rare natural gas fields containing helium at concentrations between ~0.1 and 0.3%.

Helium is used in many cryogenic applications, most notably as a liquid in the cooling of superconducting magnets used in medical MRI scanners, but is also used for pressurizing and gas purging in many industrial applications, controlled atmospheres, specialist welding, leak detection, and breathing mixtures. In 2006, total global proven reserves and probable resources were estimated to be 51.9 billion standard cubic meters. In 2014, global helium production/use was estimated to be 180 million standard cubic meters (USGS *2015*).

Neon **:** Discovered in air by Ramsay and Travers in 1898, the main source of commercial neon today is through cryogenic fractional distillation of liquid air. Best known for its use as a lighting

source in neon discharge tubes, it is also a vital component in excimer laser gas mixes used by semiconductor manufacturers. Semiconductor lithography applications using excimer lasers account for 70% of neon demand with global production in 2014 at ~400 million liters liquid neon (Corbett et al. *2015*).

Argon **:** Cavendish first observed argon in 1785 as a residual gas after removing all chemically active species from air. It was later confirmed as a component of air and subsequently named by Ramsey and Rayleigh in 1894. Like neon, the main source of commercial argon today is obtained through cryogenic fractional distillation of liquid air. This process produces ~700,000 tonnes of argon globally each year (2001). Argon is used as an inert blanket gas in the microchip and food industries and in cryogenics when the less expensive but more reactive nitrogen is not an appropriate coolant. Additional uses of argon include laser applications, to generate high energy plasma and – in liquid phase – as a scintillation target for neutrino and dark matter detectors (Acosta-Kane et al. *2008*).

Krypton **:** Discovered in 1898 by Ramsay and Travers in air, commercial krypton is also produced by cryogenic fractional distillation of air. Krypton gas finds use in lasers, as a filling for double glazing, incandescent lights, and specialist cryogenic uses as a liquid.

Xenon **:** Also discovered in 1898 by Ramsay and Travers. Commercial xenon is produced by cryogenic fractional distillation of air. Global production is about 10,000 m^3 at standard pressure and temperature (6 tonnes) of which 15% is used in medical applications as an anesthetic. In addition, xenon gas is used in modern flash bulbs; in the liquid form, it is used as a supercritical solvent, as a scintillation target for dark matter detectors, and as fuel for ion propulsion engines used in satellite positioning (Royal Society of Chemistry *2016*).

Cosmochemistry

The isotopic compositions and abundances of the noble gases in the Sun are referred to as the solar composition. This provides the reference point for chemically differentiating between meteoritic, cometary, and planetary materials. The best analog for the solar composition is solar wind, and its noble gas composition has been successfully determined by the recent NASA Genesis return mission , which trapped solar wind implanted into a variety of target materials (Wiens et al. *2007*; Meshik et al. *2014*; Crowther and Gilmour *2013*) and returned them to Earth for analysis.

The observed differences between solar and terrestrial compositions are used to identify the various origins and processes that trap volatile elements during meteorite and planetary formation (Marty *2012*; Halliday *2013*). Some noble gases are produced by radioactive decay and

cosmogenic reactions; excess abundances in these isotopes are used to provide temporal constraints on these trapping and formation processes.

Similar to other highly volatile elements such as nitrogen and carbon, the noble gas composition of the most primitive meteorites and carbonaceous chondrites are depleted by several orders of magnitude relative to the solar abundance of silicon; solar volatiles are not efficiently captured in rock-forming bodies. Superimposed upon this general depletion, light noble gases are more depleted than heavy gases (He < Ne < Ar < Kr < Xe). Furthermore, lighter isotopes of the same element are more depleted than heavier isotopes relative to the solar reference (Huss et al. *1996*; Busemann et al. *2000*; Ott *2014*).

Noble gases in meteorites are trapped in different solid phases. For example, most of the Ar, Kr, and Xe found in meteorites are found in a carbonaceous residue that remains after Hf/HCl acid treatment (i.e., "Q-phase"). In addition to Q-phase gases, noble gases are found in silicon, carbon, graphite, and nano-diamonds formed before the collapse of the solar nebula. The latter "pre-solar grains" provide isotopic compositions of Kr and Xe that are formed by s-, r-, and p-nucleosynthetic processes occurring in stars and supernovae (Ott *2014*).

In addition to Q-phase and pre-solar noble gases, significant sources of noble gas, particularly He and Ne, include ion implantation of solar wind and noble gases produced by the high energy impact of cosmogenic rays. The latter provide a crucial temporal history of meteorite breakup exposing fresh surfaces (Wieler et al. *2016*) and the travel time of Martian meteorites between ejection from Mars and impact on Earth (Nyquist et al. *2001*).

Cometary activity and meteorite breakup deliver ^{3}He-rich material to Earth in the form of interplanetary dust particles (IDPs) . These provide a significant cosmogenic flux to Earth, as IDPs retain solar wind implanted ^{3}He, which can be resolved in ocean floor sediments. Over periods of known IDP deposition, ^{3}He concentrations provide a measure of sedimentation rate. In areas of known sediment accumulation, variance in IDP flux can be attributed to, for example, high levels of cometary activity (McGee and Mukhopadhyay *2013*).

Mantle Geochemistry

Three dominant sources of noble gases exist in the Earth's mantle: (i) those trapped during the accretionary process, (ii) noble gases recycled from the planetary surface into the mantle, and (iii) those produced by the decay of extinct (^{244}Pu and ^{129}I) and extant radionuclides (U, Th, and K) within the mantle. The first work on mantle materials soon identified a dichotomy; all samples from mid-ocean ridges exhibit a very narrow range in ^{3}He/ ^{4}He (7–9 R_A) (where R_A = the ^{3}He/ ^{4}He of air = 1.4×10^{-6}), whereas most plume-influenced samples from intraplate volcanics have

$^{3}He/^{4}He$ values significantly higher (>9 R_A) and some lower than those from the mid-ocean ridges. Due to the fact that ^{3}He is dominantly sourced during accretion and ^{4}He through the radioactive decay of U and Th, this ratio provides a proxy for $^{3}He/U$ ratios in different mantle domains. The resulting layered mantle model dominated the community understanding of mantle structure and evolution for a quarter of a century, a mantle in which a well-mixed upper mantle, above the 660 km seismic discontinuity, was lithophile depleted (see lithophiles) and degassed compared to a less depleted, heterogeneous, and more volatile rich lower mantle (Kurz et al. *1982*; Allègre et al. *1983*; O'Nions and Oxburgh *1983*; Moreira *2013*). Later improvements in seismic observation of the mantle and numerical simulation of mantle convection found significant mass transfer between upper and lower mantle and a far more complex picture than the two layered mantle model (Van Der Hilst et al. *1997*; Van Keken et al. *2002*).

The Ne isotopic composition of mid-ocean ridge source mantle is indistinguishable from the solar-wind-irradiated trapped component found in meteorites (Trieloff et al. *2002*; Ballentine et al. *2005*; Ballentine and Holland *2008*). In contrast, there is some evidence in plume-derived Ne isotopes of a solar nebular signature (Yokochi and Marty *2004*; Mukhopadhyay *2012*), although this interpretation remains contested (Moreira *2013*). Similarly, the primordial MORB-source mantles Kr and Xe are also consistent with delivery of a trapped component within accreting material (Holland et al. *2009*; Caracausi et al. *2016*). This result precludes models in which the dominant sources of volatiles in Earth's mantle are derived from equilibration of an early magma ocean with a massive early (solar) atmosphere.

Xe isotopic values approaching solar values have nevertheless been found in some plume-derived samples, and this may suggest an early accretionary role for solar nebula gases (Yokochi and Marty *2004*; Mukhopadhyay *2012*). Because ^{129}I decays to ^{129}Xe ($t_{½}$ = 15.7 Ma), $^{129}Xe/^{130}Xe$ isotopic compositions can be used to infer source I/Xe compositions in the earliest Earth history. Results from the Iceland plume show that volatiles from this source have lower I/Xe than mid-ocean ridge samples, and that this signature has been preserved since ^{129}I became extinct in the first ~100 Ma of Earth history. This data is also used to argue that mixing of noble gases from the Iceland plume mantle source with mid-ocean ridge sourced volatiles does not occur at any significant level (Mukhopadhyay *2012*).

The primordial heavy noble gases in the mid-ocean ridge basalt source mantle are accompanied by atmosphere noble gases with an elemental composition consistent with subduction of an ocean/pore-fluid signature into the mantle (Holland and Ballentine *2006*). This oceanic pore-fluid signature is observed in both halogens and noble gases in fluids subducted to ~100 km depth (Sumino et al. *2010*), demonstrating preservation of the pore-fluid signal to depths at which transfer into the deeper mantle can be envisaged. Linking subduction flux of the noble gases to

volatile species such as carbon and nitrogen (Marty and Jambon *1987*; Barry and Hilton *2016*) has provided a powerful tool to trace major volatile subduction into the mantle (Hilton et al. *2002*).

The location and processes, recorded by the noble gases, that enable preservation of different accretionary volatile components in the mantle since the formation of Earth and the superposition of radiogenic and recycled noble gases remain at the frontier of the current research effort.

Earth's Atmosphere

The noble gas composition of Earth's atmosphere is well mixed and provides the reference abundance and isotopic composition for most noble gas terrestrial studies (e.g., Sano et al. *2013*). With the exception of ^{40}Ar and He, the atmosphere contains >98–99% of all terrestrial noble gases. The pattern of the non-radiogenic noble gas isotopes is, similar to chondrites, strongly depleted in both light elements and light isotopes relative to solar. Xe abundances are, however, depleted by an order of magnitude more than would be expected using the chondritic pattern as a reference, and very strongly isotopically fractionated. This is often referred to as the "missing xenon" paradox (Marty *2012*).

Xe isotopes derived from the extinct radioisotopes ^{129}I and ^{244}Pu can be resolved in the atmosphere. These are highly depleted relative to predicted amounts and are used to argue that the earliest atmosphere was not retained until "closure" at ~40–100 Ma after the Earth formed (Pepin and Porcelli *2006*; Avice and Marty *2014*). The proportion of radiogenic noble gas isotopes from extant radionuclides has increased in the atmosphere over geological time due to Earth degassing. This is most notably seen in ^{40}Ar relative to ^{36}Ar and is a key measure of mantle outgassing efficiency and continental crustal growth (Allègre et al. *1996*; Pujol et al. *2013*). Analysis of fluids trapped in ancient minerals and free fracture fluids shows a light isotope enrichment in shielded Xe isotopes (^{124,126,128}Xe) (Pujol et al. *2011*; Holland et al. *2013*). It is argued that this is evidence of xenon loss from Earth's atmosphere that provides one explanation for the missing xenon (Pujol et al. *2011*; Marty *2012*).

Over the last 1Ga, changes in the isotopic composition and partial pressure of the noble gases in the terrestrial atmosphere have been small and implicitly assumed negligible in work using the modern atmosphere as a reference.

Ice, Oceans, and Crustal Fluids

The physical processes of diffusion and solubility which fix atmosphere-derived noble gases into ice and water are well understood. Small differences in environmental conditions such as

temperature and salinity provide measurable changes in the noble gas content of ice and water, preserving temporal records of environmental change.

Transient temperature gradients fractionate air trapped within firn through thermal diffusion which is then preserved in air bubbles in ice. Ice cores therefore provide a temporal record of both temperature change and rate of change (Severinghaus and Brook *1999*; Winckler and Severinghaus *2013*).

Similarly, when water equilibrates with the atmosphere, the concentration of noble gases in the water phase is determined by the temperature and salinity of the water as well as the partial pressure of the noble gases. Dynamical features of air dissolving into water can introduce nonequilibrium air concentrations into the water, but these are reasonably well understood.

In ocean environments, noble gas concentrations that deviate from equilibrium are used to diagnose and quantify ice formation, melting, and/or bubble injection through increased wave activity. Argon can be used to provide a baseline for N_2 and O_2 activity, allowing differentiation between physical and biogeochemical activity and thus identification of the processes controlling biologic production and denitrification.

Non-atmosphere input occurs in the form of ^{3}He from mid-ocean ridges and atmosphere tritium decay ($t_{1/2}$ = 12.3 years). The former provides a tool for deriving basin-scale mixing, global model verification, and trace element addition to different ocean bodies through hydrothermal activity. The latter provides information about the age and extent of recent surface water incursion (Schlosser and Winckler *2002*; Stanley and Jenkins *2013*).

On the continents for a known location (altitude) and freshwater recharging an aquifer (near zero salinity), local temperatures at recharge can typically be determined from the noble gas concentrations in water to better than ±0.3–0.5 °C (Ballentine and Hall *1999*). With knowledge of the mean residence time of the water in a given aquifer, changes in the local paleoclimate can be reconstructed (Kipfer et al. *2002*; Aeschbach-Hertig and Solomon *2013*).

Like ocean water, tritium- ^{3}He can be used to provide a mean residence time (MRT) for young (<50 years) groundwaters. ^{85}Kr ($t_{1/2}$ = 10.76 years) released into the atmosphere by anthropogenic nuclear activity has similar potential but requires large sample volumes and specialized atom trap trace analysis (ATTA) techniques. Cosmogenically produced ^{39}Ar ($t_{1/2}$ = 269 years) is formed in the atmosphere and has the potential to significantly extend the capability to trace young groundwater. It also requires ATTA technology for analytical determination; however, recent advances in the technique has allowed for smaller sample quantities to be accurately measured (Yang et al. *2013*; Ritterbusch et al. *2014*).

With the development of ATTA, cosmogenic ^{81}Kr can now also be determined in groundwater and ice. With a half-life of 229,000 years., ^{81}Kr extends the ability to date water MRTs and ice

core ages beyond the limit of ^{14}C dating to over ~1 Ma (Aggarwal et al. *2015*). For fluids with MRTs >1 Ma, the radiogenic noble gases (^{4}He, $^{21,22}Ne$, ^{40}Ar, $^{131,132,134,136}Xe$) can be used if the accumulation rate is known (Ballentine and Burnard *2002*). For a closed system which releases all noble gases into the fluid phase, this is given by the concentration of radioelements and has been used to identify fluids in the crust >1 Ga (Holland et al. *2013*). When an aquifer is open to flux into the system, the external flux rate needs to be determined; this may be from local proximal formations with high radioelement concentrations (Tolstikhin et al. *2011*) or by using flux estimates based on the steady-state release of radiogenic noble gases from the whole crust (Torgersen *2010*).

When a second fluid phase such as natural gas, oil, or carbon dioxide passes through the groundwater system, the noble gases equilibrate between phases as a function of local conditions (temperature, pressure, and salinity) and fluid volume ratio, and/or whether the process is open or closed system (Zhou et al. *2005*; Barry et al. *2016*). In well-behaved systems, the resulting noble gas content of either phase preserves a quantitative record of this phase interaction. This has been used to identify the role of groundwater in oil and gas migration to trap and identify sources and sinks of non-hydrocarbon gases such as CO_2, N_2, and commercial helium accumulation (Ballentine et al. *2002*; Gilfillan et al. *2009*; Prinzhofer *2013*, Byrne et al. *2016*). Similar principles apply in hydrothermal systems where boiling or other phase separation processes can be traced (Sano and Fischer *2013*) and their role in mineral deposition investigated (Kendrick et al. *2001*).

Geochronology

Noble gas accumulation rates within minerals provide a wealth of information for geochronological applications throughout cosmochemistry and the Earth sciences. Arguably the largest impact has been the exploitation of K decay to ^{40}Ar ($t_{½}$ = 1250 Ma). Early development of K-Ar was very rapidly superseded by reactor neutron conversion of K to ^{39}Ar, with simultaneous measurement of ^{40}Ar- ^{39}Ar providing accuracy and a temporal range applicable “from archaeology to planetary sciences” (Kelley *2002*; Jourdan et al. *2016*).

A related offshoot to ^{40}Ar- ^{39}Ar chronology is the conversion of halogens (F, Cl, Br, I) by neutron reaction to noble gas isotopes, providing a powerful analytical tool for halogen determination in samples where other techniques (particularly for Br and I) do not provide the necessary sensitivity (e.g., Ruzié-Hamilton *2016*).

Cosmogenic nuclides provide surface exposure ages on meteorites and terrestrial surfaces alike (Niedermann *2002*; Dunai et al. *2007*; Wieler et al. *2016*), while (U-Th)/He takes advantage of the low closure temperature in minerals (e.g., apatite) and provides a thermochronological history that

can delineate the cooling history of rocks in a temperature range unobtainable by other techniques (Farley *2002*, Reiners et al. *2004*) .

Summary and Conclusions

Noble gases have a wide array of terrestrial geochemical applications in addition to great utility in cosmochemistry. Due to the unique sources of different isotope species, the noble gases can be readily exploited to differentiate between atmospheric, mantle, crustal, and extraterrestrial volatile sources. Furthermore, radiogenic noble gas species can be used to provide temporal constraints on the timing of geologic closure events. Together, noble gases form a unique and extremely diverse set of geochemical tracers that have far-reaching applications in Earth and planetary sciences.

Cross-References

Chronology

Cosmogenic Nuclides

Crustal Fluids

Earth's Ccontinental Crust

Earth's Mantle

Oil, Water, Gas

Periodic Table

Radon

Solar System Formation

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