Geochemical Perspectives Letters

Sun flare activity may solve unknown source of helium-3 in the atmosphere

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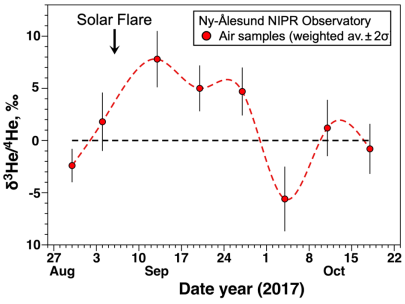
https://doi.org/10.7185/geochemlet.2237

Abstract



Helium abundance measurements in the atmosphere suggest a supply of crustal ^{4}He from natural gas extraction since the beginning of the 20th century. However, the ^{3}He/^{4}He ratio of air appears near constant, which calls for an unknown source of ^{3}He to compensate for the contribution of anthropogenic ^{4}He to the atmosphere. Knowing the origin and cycle of ^{3}He in the atmosphere is important, being also an important resource in nuclear fusion reactors and for cryogenic applications. Here, we report ^{3}He/^{4}He variations measured during the massive X9.3 solar flare event of September 6th 2017 in Ny-Ålesund, Svalbard islands, near the North Pole. The solar wind was expected to reach the Earth on September 8th. A total of five samples, collected immediately after the solar flare event, showed an excess of ^{3}He, up to 5.5 ± 1.7 ‰ (δ^3He), compared to the terrestrial atmospheric isotopic value. If the solar wind, enhanced by solar flares, was fed into the atmosphere by auroral precipitation, it would increase the polar atmospheric helium isotope ratio. The helium would then be diluted by diffusion and the excess ^{3}He would rapidly disappear. Thus, ^{3}He excess supplied by these events may keep the atmospheric ^{3}He/^{4}He constant.

Received 18 June 2022 | Accepted 14 September 2022 | Published 18 October 2022

Introduction

Helium concentration in the atmosphere is determined by the balance between degassing from the solid Earth and the thermal and/or non-thermal escape from the upper atmosphere (MacDonald, 1963). However, accelerated extraction and consumption of fossil fuels since the beginning of the 20th century may have altered this balance. Natural gas contains about 0.25 % helium (Zartman *et al.*, 1961) with an isotopic ratio (Aldrich and Nier, 1948) much smaller than that of the atmospheric value (1.382×10^{-6}; Sano *et al.*, 2013), because of the dominance of crustal ^{4}He compared to ^{3}He in fossil fuels. If a large amount of helium from extracted natural gas is added to the atmosphere, the ^{4}He concentration in air should increase with time, while the isotope ratio ^{3}He/^{4}He should decrease. Oliver *et al.* (1984) first reported a rough estimate of the ^{4}He released in the atmosphere due to natural gas production ($3–12 \times 10^{16}$ cm^3 STP between 1939 and 1981). Sano *et al.* (1989) measured a temporal decrease in the helium isotope ratio in the atmosphere of ~1 x 10^{-9}/year. Although there has been much debate since then, recent analyses of the ^{3}He/^{4}He ratio in sampled air of different epochs have concluded that the atmospheric ^{3}He/^{4}He ratio is rather constant (Lupton and Evans, 2004; Mabry *et al.*, 2015; Boucher *et al.*, 2018). Recently, Birner *et al.* (2022) developed a method to measure helium concentrations with high precision (±0.07 ‰) and analysed atmospheric samples from the past 46 years. The results showed an increase in ^{4}He concentration of 1.9 ‰ and these authors concluded that the increase was due to anthropogenic release of helium from natural gas. This apparent discrepancy – *i.e.* constant ^{3}He/^{4}He ratio but contemporary increase of anthropogenically-released ^{4}He – suggests that an unknown source of ^{3}He is compensating for the addition of crustal ^{4}He (Birner *et al.*, 2022). This unknown source of ^{3}He is here defined as the "missing helium-3". Knowing the origin and fate of this source of helium is important, given that ^{3}He is an important but scarce resource on Earth, necessary for the development and operation of nuclear fusion reactors and for cryogenic applications. Based on changes in the atmospheric helium isotope ratios measured in polar regions, the present study suggests that solar flares may be the source that supplies the ^{3}He required to account for the near-constant ^{3}He/^{4}He atmospheric ratio.

Samples

Atmospheric samples used in this study were collected at Ny-Ålesund (78°55'N, 11°56'E), Svalbard islands, near the North Pole, in the Norwegian-controlled International Observation

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Village. At Ny-Ålesund, atmospheric samples are usually collected weekly by collaborators at the Norwegian Polar Institute and transported to Japan approximately every two months. Each sample is collected using a diaphragm pump from an air intake on the roof of the Japanese base in Ny-Ålesund, passed through a water trap cooled to −78 °C, and pressurised into a pre-evacuated 800 ml stainless steel container. On September 6th, 2017, a massive solar explosion, or solar flare, occurred. The solar wind was expected to reach the Earth on September 8th (NASA, 2017). Therefore, the dates of air sample collection were set for eight days about one week apart, from August 30th to October 18th, 2017. These samples were rapidly airlifted from Ny-Ålesund to Japan. At the end of October 2017, the atmospheric samples preserved in metal cylinders stored at the National Institute of Polar Research were transferred into lead glass reservoirs in Tachikawa City, Tokyo. The samples were brought to the Atmosphere and Ocean Research Institute of The University of Tokyo (Kashiwa, Chiba) and analysed for helium isotope ratios in May 2018.

Analytical Procedures and Results

Noble gases contained in the samples were purified by an activated charcoal trap that was cooled to liquid nitrogen temperature together with a heated titanium getter. Finally, helium was separated and purified from neon in a cryogenic trap containing activated charcoal cooled to 40 K. Helium isotope ratios were measured with a double-collector noble gas mass spectrometer, GV® Helix SFT (Sano *et al.*, 2008). Measurements were performed 2–3 times for each individual sample and compared to a synthesised helium standard, HESJ (Matsuda *et al.*, 2002) with an accepted $^3He/^4He$ value of 20.63 ± 0.10 Ra. The measured $^3He/^4He$ ratios were then normalised to the standard value of the atmosphere, 1.382×10^{-6} (Sano *et al.*, 2013).

In order to determine the atmospheric helium isotopic background at the Ny-Ålesund site, air samples were collected from January 2020 to January 2021, about three months apart, during a quiescent period of solar activity. These data are presented in Table S-1 and the $^3He/^4He$ variations are shown in Figure 1. The $^3He/^4He$ ratios vary from −3.0 ‰ to + 3.0 ‰, consistent with our laboratory air standard collected in the Kashiwa Park (Latitude, 35.8949; longitude, 139.9415) close to the campus of University of Tokyo, within experimental error. The weighted mean and internal error on 15 measurements are −0.47 ± 0.79 (see Supplementary Information for calculation), not distinguishable from the air value, assuming uncertainties. Therefore, there is no anomaly of the atmospheric helium isotopic ratio during the January 2020 to January 2021 period and the analytical system can be considered significantly stable.

The helium isotopic ratios of analysed Arctic air during a solar flare event are shown in Table S-2 and reported as δ values (deviation from the Kashiwa air standard expressed in part per thousand). Three runs were made on the same sample, with the second run being the most stable; for the first and third runs, some of the data were erroneous due to instrumental fluctuations and are not included in the discussion due to large uncertainty. The $^3He/^4He$ ratios of Arctic air varied from −5.6 ± 3.1 ‰ to +8.4 ± 3.4 ‰ (Table S-2). The $^3He/^4He$ ratios of 12 of the 18 samples agreed with the atmospheric values of Kashiwa air standard, within the error range. Of the six data showing anomalies, the five collected on September 13th, 20th, and 27th of 2017 showed a clear excess of ^3He compared with the atmospheric ratio (Fig. 2). These data are noteworthy because they were taken immediately after the massive solar flare event of September 6th that registered X9.3 on the solar storm scale, which divides solar flares according to their strength (NASA, 2017). The smallest events are A-class (near background levels), followed by B, C, M and X, with each letter class having a finer scale from 1 to 9, except class X which can go beyond it (with X28, detected in 2003, being the highest solar flare ever detected). Events equivalent and larger than X9.3 have occurred 27 times from 1975 to 2018 (NIICT, 2018), so their occurrence is not significantly unusual. The other $^3He/^4He$ data are consistent with the standard atmospheric ratio, except for the data from the first run on October 4th, which shows a ^3He deficiency for an unknown reason. We therefore propose that the source of the "missing helium-3" excess is the injection of solar wind-derived ^3He into the atmosphere, enhanced by the solar flare as shown in

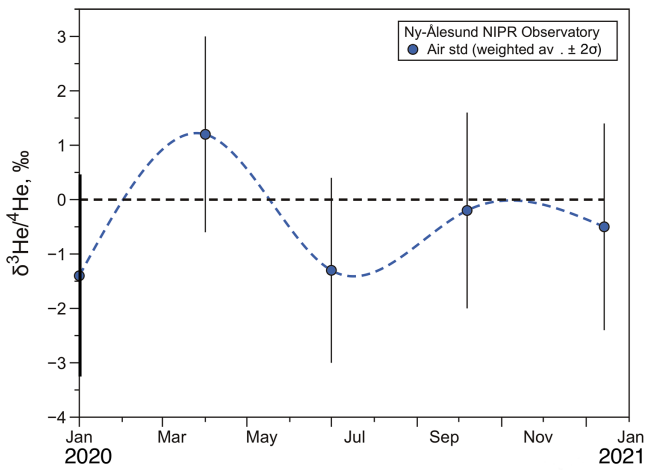

Figure 1 Temporal variation of atmospheric $^3He/^4He$ weighted averages in Ny-Ålesund, Svalbard, near the North Pole, from January 2020 to January 2021. The black dotted line and blue dotted curve show the standard atmospheric helium value and a spline curve approximating the time variability of the weighted average $^3He/^4He$ ratio for each sampling day, respectively. Bars indicate 2σ internal errors (see Supplementary Information for equations).

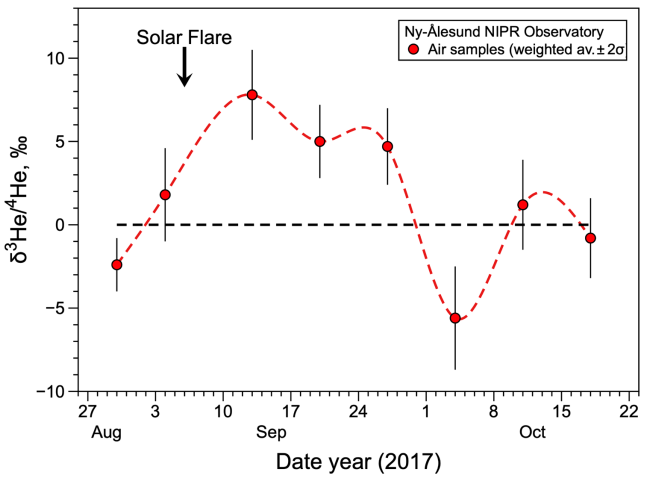

Figure 2 Temporal variation of atmospheric $^3He/^4He$ weighted averages in Ny-Ålesund, Svalbard, near the North Pole, around the time of the X9.3 solar flare event. The black dotted line and red dotted curve show the standard atmospheric helium value and a spline curve approximating the time variability of the weighted average $^3He/^4He$ ratio for each sampling day, respectively. Bars indicate 2σ internal errors (see Supplementary Information for equations).

Figure 2. This study illustrates for the first time the detection of a local excess of ^{3}He in the lower atmosphere due to a solar flare.

Discussion

The atmospheric ^{3}He concentration should be 7.24 pptv, based on the helium concentration in the atmosphere of 5.24 ppmv (Gluckauf and Paneth, 1946) and a ^{3}He/^{4}He of 1.382×10^{-6} (Sano *et al.*, 2013). A dominant source of atmospheric ^{3}He is the Earth's mantle through volcanic activity and degassing from mid-oceanic ridges (Clarke *et al.*, 1969) and subduction zones (Sano and Wakita, 1985). Another source is the interplanetary zone (Ozima and Podosek, 1983). When an explosive event (solar flare) occurs in the sun, a plasma plume (solar wind) containing a large amount of ^{3}He is generated and approaches the Earth in a path along the magnetic field lines. This ^{3}He is captured as it passes through the Earth's magnetosphere and is precipitating into the upper atmosphere of the polar regions. This air, locally mixed with air downward, may tentatively produce a high ^{3}He/^{4}He ratio. This mechanism is associated with the formation of auroras and is called Auroral Helium Precipitation (Buhler *et al.*, 1976), and it may explain excess ^{3}He observed in this work. Other sources include the decay of tritium produced by nuclear weapons and nuclear reactors, but it is thought to be negligible in amount (Lupton and Evans, 2004). If the solar wind, enhanced by solar flares, were fed into the atmosphere by the auroral precipitation mechanism, it would increase the polar atmospheric helium isotope ratio, as shown in Figure 2. The helium would then be diluted by diffusion and the excess ^{3}He would disappear soon after. Mixing of atmosphere along the latitude from east to west may take a few weeks by air transport, which is much faster than that of a few months along the longitude from north to south, generally observed by long-lived tracers (Jacob, 1999). In the present case, the lifetime of excess ^{3}He may be short when compared with the mixing time. This discrepancy may be resolved by analysing more case studies of solar flare events. In addition to horizontal mixing of the atmosphere, vertical mixing must also be considered. Experiments using rockets have observed ^{3}He of solar wind origin at an altitude of about 150 km (Axford *et al.*, 1972). However, there is no report on the mixing mechanism from the thermosphere to the ground surface. According to turbulent diffusion models, vertical mixing between the troposphere and the surface takes more than one month (Jacob, 1999). In our observations, the time difference between a solar flare observation and an increase of ^{3}He/^{4}He in the atmosphere is only a few days (Fig. 2). Thus, the change in helium is faster than the model mixing. It is beyond the scope of this paper to provide a physical explanation for the cause of the rate being too fast and the reason will be clarified in future studies. In addition, the surface region most strongly influenced by the stratosphere is found substantially south of the sampling location (Škerlak *et al.*, 2014). The ^{3}He signals originating from the stratosphere should therefore be larger at lower latitudes and we might underestimate the effect of auroral precipitation in Svalbard.

To test the hypothesis of downward injection of solar ^{3}He, a discussion on the mass balance of ^{3}He is necessary. The observed increase in atmospheric helium concentration (Birner *et al.*, 2022) suggests an anthropogenic supply of crustal ^{4}He of $3.9 \pm 0.3 \times 10^{10}$ mol/year (2σ). Birner *et al.* (2022) calculated that to compensate for the addition of anthropogenic ^{4}He and to preserve a constant atmospheric ^{3}He/^{4}He ratio, the annual addition of $6.3 \pm 2.5 \times 10^4$ mol of ^{3}He is required. This is equivalent to an annual change in the atmospheric helium isotope ratio of $+0.049 \pm 0.020$ ‰ (Birner *et al.*, 2022). However, this calculation requires that the ^{3}He/^{4}He of natural gases is $1.63 \pm 0.68 \times 10^{-6}$, which is much higher than the typical crustal ratio of 2×10^{-8}, and it can be found only in hydrocarbons settled in extensional sedimentary basins where crustal and mantle helium can be found together (*e.g.*, Ballentine *et al.*, 1991). Since natural gas and oil are often formed in marine sediments, we may assume that the helium isotope ratio of natural gas is $4.5 \pm 3.5 \times 10^{-7}$, the average value of marine sediment pore water measured in the ocean floor (Sano and Wakita, 1987) and consistent with early data in natural gases (Aldrich and Nier, 1948). It leads to the required isotope ratio change of $+0.028 \pm 0.022$ ‰/year, comparable with the lower figure estimated by Birner *et al.* (2022). Thus, the helium isotope variation to be adjusted and offset would be between 0.028 ± 0.020 ‰/year and 0.049 ± 0.020 ‰/year.

The total amount of ^{3}He injected by solar flares can be estimated based on the variation of the atmospheric helium isotope ratio, illustrated in Figure 2. The positive helium isotope ratio anomaly lasted from September 13th to September 27th with an average deviation of 5.5 ± 1.7 ‰ calculated from the six anomalous samples. The area of the Earth corresponding to the polar regions where auroral precipitation is effective is estimated to be about 3 % of the total area of the Earth (Buhler *et al.*, 1976). If the 5.5 ‰ ^{3}He excess is globally diffused and diluted, then the ^{3}He/^{4}He ratio of the entire atmosphere would increase by 0.165 ‰ by only one large event. Based on the observations, there were 27 events larger than X9.0 from 1975 to 2018 (NIICT, 2018). This suggests almost one solar flare event *per* two years. If these events have similar effect on the atmosphere, the increase in the air helium isotope ratio would be 0.10 ± 0.03 ‰/year on average, which is much larger than the range of 0.028–0.049 (±0.020) ‰/year calculated and predicted above. Thus, large solar flare events could supply sufficient ^{3}He in the atmosphere to compensate for the addition of anthropogenic ^{4}He, even if diluted by the complex atmospheric circulation processes described above.

If the increase in the helium isotope ratio due to the solar flare on September 6th is 0.165 ‰ on an annual basis, based on the atmospheric inventory of ^{3}He, *i.e.* 1.5×10^{14} atoms/cm^2 (Ozima and Podosek, 1983), it would result in a flux of 190 atoms/cm^{2}sec. This value is about 40 times greater than the steady state auroral precipitation flux of 5 atoms/cm^{2}sec (Buhler *et al.*, 1976). In other words, even if a solar flare of this scale were to occur once every two years, it would be 20 times larger than the normal auroral precipitation flux. Therefore, solar flares would be an important source controlling atmospheric ^{3}He concentration, even if over estimated, compensating for its decrease caused by the injection of crustal ^{4}He from natural gas extraction and consumption by humans.

Conclusions

In order to maintain the ^{3}He/^{4}He ratio of the atmosphere constant despite an increasing contribution of crustal ^{4}He during the last century caused by extraction of natural gas, the injection of solar flare ^{3}He should also have increased during a comparable period of time. The record of solar activity based on archival proxies, such as the concentration of the cosmogenic isotopes ^{14}C in tree rings (Solanki *et al.*, 2004) or ^{10}Be in ice cores (Beer, 2000), may provide good examples, although other sources of cosmogenic isotope fluctuations are also possible (Heaton *et al.*, 2021). Based on the precise measurements of air helium isotopes in a polar region, we found the first case of temporal variation and claim that sun flare activity may alter global mass balance of ^{3}He in the atmosphere.

Acknowledgements

We are grateful to the Norwegian Polar Institute's staff for their careful air sampling at Ny-Ålesund. Thanks also for helpful comments from two anonymous reviewers.

Editor: Maud Boyet

Additional Information

Supplementary Information accompanies this letter at https://www.geochemicalperspectivesletters.org/article2237.

Cite this letter as: Sano, Y., Pinti, D.L., Escobar-Nakajima, T., Takahata, N., Zhang, M., Goto, D., Marty, B. (2022) Sun flare activity may solve unknown source of helium-3 in the atmosphere. *Geochem. Persp. Let.* 23, 49–52. https://doi.org/10.7185/geochemlet.2237

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