



Precise measurement of atmospheric helium isotopes

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Several recent papers have addressed the question of whether the atmospheric $^3\text{He}/^4\text{He}$ ratio is decreasing with time. Sano et al. measured the $^3\text{He}/^4\text{He}$ ratio of air samples collected over a 10-year period at various sites in Japan and claimed an apparent 1-2% decrease in the ratio. Lupton and Graham reported a slope of $+0.009\pm 0.043$ %/year (2σ) for the time rate of atmospheric $^3\text{He}/^4\text{He}$ change (TROC) based on the Pacific marine air collected over a 17-year period. Pierson-Wickman et al. measured $^3\text{He}/^4\text{He}$ ratios of air trapped in historical metallurgical slag in an attempt to study secular variation of the ratio. Taking into account of data, they proposed a TROC of -0.014 ± 0.007 %/year. Recently Lupton and Evans reported that a TROC could be between -0.0102 and $+0.0019$ %/year, which corresponds to a 0.3% decrease at the most in the air $^3\text{He}/^4\text{He}$ over the past 3 decades. Even though many efforts have been devoted to secular variation of air helium, the change of atmospheric $^3\text{He}/^4\text{He}$ ratio with latitude and longitude have not been assessed since the pioneer work of Mamyrin et al. We have developed an analytical system to measure atmospheric $^3\text{He}/^4\text{He}$ ratios precisely by using a static vacuum operation mass spectrometer and an ultrahigh-vacuum purification line. Repeated analysis of a sample calibrated against a standard shows the precision of about 0.2% error margin (2σ). This system has been applied to the evaluation of $^3\text{He}/^4\text{He}$ ratio of He Standard of Japan (HESJ), and to the assessment of atmospheric variation of latitude. Observed $^3\text{He}/^4\text{He}$ ratio of HESJ, 20.405 ± 0.040 R_{air} (2σ) agrees well with the value of 20.408 ± 0.044 R_{air} by Lupton and Evans, but is smaller than the recommended value of 20.63 ± 0.10 R_{air} in a literature. Air samples collected from 45°N to 20°N in the Far East show a slight decrease of $^3\text{He}/^4\text{He}$ ratio with latitude. Even though the variation is attributable to an experimental artifact, it may not preclude the possibility that the anthropogenic release of crustal ^4He

is significant in the high latitude region of the northern hemisphere.