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Origin of nitrous oxide dissolved in the ocean deduced from concentration and isotope ratios

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Nitrous oxide (N_2O) is a trace gas that contributes to both global warming and stratospheric ozone depletion. Its major sources are nitrification and denitrification by microorganisms in soils and waters. Emission of N_2O from the ocean to the atmosphere is estimated to take account for 20% of global N_2O sources. Based on the distribution of N_2O dissolved in the surface seawater, most of the emission is considered to occur near the eutrophic, suboxic water mass like eastern tropical Pacific, where N_2O is produced by denitrification. However, N_2O concentration shows its maximum in middle layer (600-1000 m) in many sea areas, and the origin of the concentration maximum is not clear. In addition, N_2O is also supersaturated in deep water and its concentration has been found to be different among sea areas. In this study, we focus attention on isotope ratios of N_2O (oxygen isotope ratio, delta-^{18}O, difference in abundance of isotopomers $^{14}N^{15}N^{16}O$ and $^{15}N^{14}N^{16}O$, Site Preference (SP), and average nitrogen isotope ratio, delta-^{15}N) as informative parameters reflecting microbiological metabolisms, and made an analysis of origin of N_2O in the middle and deep ocean by the use of distribution of isotope ratios as well as concentration.

In addition to published data obtained in the western North Pacific, subtropical North Pacific, eastern tropical North Pacific, eastern subtropical North Pacific, and eastern subtropical South Pacific, we obtained depth profiles of N_2O and its isotope ratios in the Southern Ocean, equatorial Pacific, and North Pacific by analyzing the samples collected in cruises KH01-3, MR02-K06, and MR07-04, respectively. Comparison of the profiles indicated that (1) the dissolved N_2O concentration reaches its maximum in the layer of sigma-theta = 26.5-27.5, which corresponds to the depth of 100-300 m in the Southern Ocean and equatorial Pacific and 700-1500 m in other regions, and (2) the maximum concentration is about twice and 4-7 times as large as saturation concentration for atmosphere-ocean equilibrium in the southern (Southern Ocean and South Pacific) and other stations, respectively. Isotope ratios at the N_2O concentration maxima also shows a variation among the ocean areas, but there is no significant relationship between the concentration and isotope ratios. This means that the origin of N_2O maxima observed in the Pacific is not identical, and in situ production plays more important role than lateral advection or diffusion. Assuming that the N_2O excess is caused by in situ production, we estimated isotope ratios of the produced N_2O as 7-11 permil, 50-65 permil, and 18-35 permil for delta-^{15}N, delta-^{18}O, and SP, respectively. The SP value was higher in the northern stations than in the southern stations. On the other hand, average concentration and isotope ratios of N_2O in the deep layer (below 2000 m) show a significant correlation with the age of the water mass estimated from 14C concentration, suggesting that N_2O is produced during the deep water circulation. Estimated isotope ratios for the N_2O produced were 10 permil, 62 permil, 29 permil for delta-^{15}N, delta-^{18}O, and SP, respectively.

The estimated delta-^{15}N and SP of N_2O produced in the middle and the deep layer almost agree with the values of N_2O produced during nitrification (NH_2OH oxidation) by ammonia-oxidizing archaea (AOA), which were recently obtained by laboratory incubation experiments. However, the estimated delta-^{18}O value in the middle/deep water is about 20 permil higher than that of AOA-produced N_2O, and SP of N_2O in the middle layer shows lower value in some areas. We therefore cannot exclude the contribution from bacterial nitrification and denitrification (nitrite reduction by nitrifier or denitrifier). Further researches on isotopic fractionation during the N_2O production by AOA is needed with respect to its variation among species and dependence of environmental factors.

Keywords: Nitrous oxide, ocean, stable isotope ratio, nitrification, denitrification, ammonia-oxidizing archaea