ISOTOPOMER STUDIES OF NITROUS OXIDE IN LOW OXYGEN MARINE ENVIRONMENTS

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ABSTRACT

The oceans are a significant source of the greenhouse gas, nitrous oxide, contributing about 30% of the natural sources and 17% of total identified sources. Nitrous oxide can be produced in the oceans by nitrification, the conversion of ammonium to nitrite and nitrite to nitrate in the presence of free oxygen, and can be produced and consumed by denitrification, the oxidation of organic matter using nitrogen oxides as electron acceptors in the absence or near absence of free oxygen. The relative yield of nitrous oxide from both processes is enhanced by low oxygen conditions. In this dissertation, I investigate the dynamics of nitrous oxide in two very different low oxygen environments: the waters overlying the intense oxygen minimum zone of the eastern tropical North Pacific and the suboxic layer above the permanently anoxic Black Sea basin. In order to distinguish between nitrification and denitrification, I apply a novel form of isotope ratio monitoring mass spectrometry that allows the determination of position-dependent nitrogen isotope ratios in dissolved nitrous oxide. In the first part of my dissertation I present a solution to an ongoing controversy concerning the calibration of position-dependent nitrogen isotope ratio measurements on different mass spectrometers. I then apply this measurement to the subsurface nitrous oxide concentration maximum in the eastern tropical North Pacific and determine that the primary source of nitrous oxide in this feature is nitrifier denitrification, the conversion of nitrite to nitrous oxide during the course of ammonia oxidation. In addition, I present isotopic evidence that nitrous oxide is simultaneously produced and consumed at the depth of the subsurface concentration maximum. Measurements in the Black Sea show

simultaneous production and consumption of nitrous oxide in the suboxic zone with the net result that the gas does not accumulate at concentrations that could yield a significant flux to the atmosphere. Based on my field work, I assess the assumptions underlying current models of the oceanic nitrous oxide cycle and argue for the inclusion of a consumption term to improve simulations of the oceanic nitrous oxide distribution and estimates of total losses of fixed nitrogen from the ocean.