The North Atlantic Ocean as a source of atmospheric N_2O

By JÜRGEN HAHN, *Max-Planck-Institut für Chemie, Mainz, W.-Germany*

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ABSTRACT

In 1969, 1970, and 1971 N_2O measurements of sea water were carried out during 3 cruises in the open North Atlantic ocean. Water samples were taken from the sea's surface and from depths down to 3 000 m at several stations. Surface water concentrations in the tropical-subtropical latitudes averaged 0.5 μg N_2O per liter sea water. For the range from 38.5° N to 48.5° N, an average of 0.4 μg N_2O per liter sea water was found. In the area of the Iceland–Faroe ridge, the surface water concentrations averaged 0.4 μg N_2O per liter sea water when water temperatures were 10°C, and 0.5 μg N_2O per liter sea water when water temperatures were 5°C. The vertical N_2O concentration profiles often show two maxima: a smaller one between 100 and 200 m and a large one between 400 and 1 000 m with N_2O concentrations up to 0.8 μg per liter sea water. With the exception of a few samples, the measurements indicated that the North Atlantic sea water is supersaturated with N_2O. Supersaturation values up to more than 100 % were found in the upper layers of the sea water from the tropical-subtropical latitudes up to 48.5° N. Further north the N_2O supersaturation of the sea water decreases. For the upper sea water layers down to 1 000 m the following average values were obtained from vertical profiles: in tropical latitudes about 66 % supersaturation, in subtropical latitudes about 47 %, in the range from 38.5° N to 48.5° N about 42 %, and in the area of the Iceland–Faroe ridge about 12–20 % supersaturation. One may conclude that the North Atlantic ocean acts as a net source of atmospheric N_2O. It is probable that the other oceans have the same ability.

Introduction

When we began our study of atmospheric trace gases some years ago, it was known for more than 25 years that nitrous oxide is a regular constituent of the earth's atmosphere. Estimates and theoretical considerations based primarily on laboratory studies and a few measurements in the atmosphere pointed to soil bacteria as the major source and to photochemical destruction in the troposphere and stratosphere as the major sink for nitrous oxide.

The N_2O data we obtained from 1966 through 1969 during a comprehensive field program were discussed in a paper by Schütz et al. (1970). Using these data, the atmospheric life time of N_2O was calculated to less than 10 years, while calculations with the photochemical destruction rate given by Bates & Hays (1967) resulted in atmospheric life times of about 70 years. The conclusion we drew from this discrepancy was that there must be additional sinks for N_2O in the troposphere or hydrosphere.

The first N_2O data from sea water were measured by Craig & Gordon (1963) in the South Pacific ocean. These rather preliminary data seemed to indicate that the sea acts as a slight sink for N_2O. Reliable and more numerous data on this question were, therefore, of particular interest to us, and so we joined in 3 cruises of the German research vessel *Meteor* in 1969, 1970, and in 1971 to measure N_2O concentrations in marine air and in sea water.

Analytical method

The analytical method used for the N_2O measurements at sea was published in papers by Bock & Schütz (1968), by Junge et al. (1971), and by Hahn (1972), so that only a brief description of the method is necessary: The N_2O from an air or water sample, respectively, is adsorbed on 5A molecular sieve and then transferred for analysis to a gas chromatographic column. From water samples the N_2O is expelled by heating and passing through a stream of clean nitrogen. During the 1969 Expedition

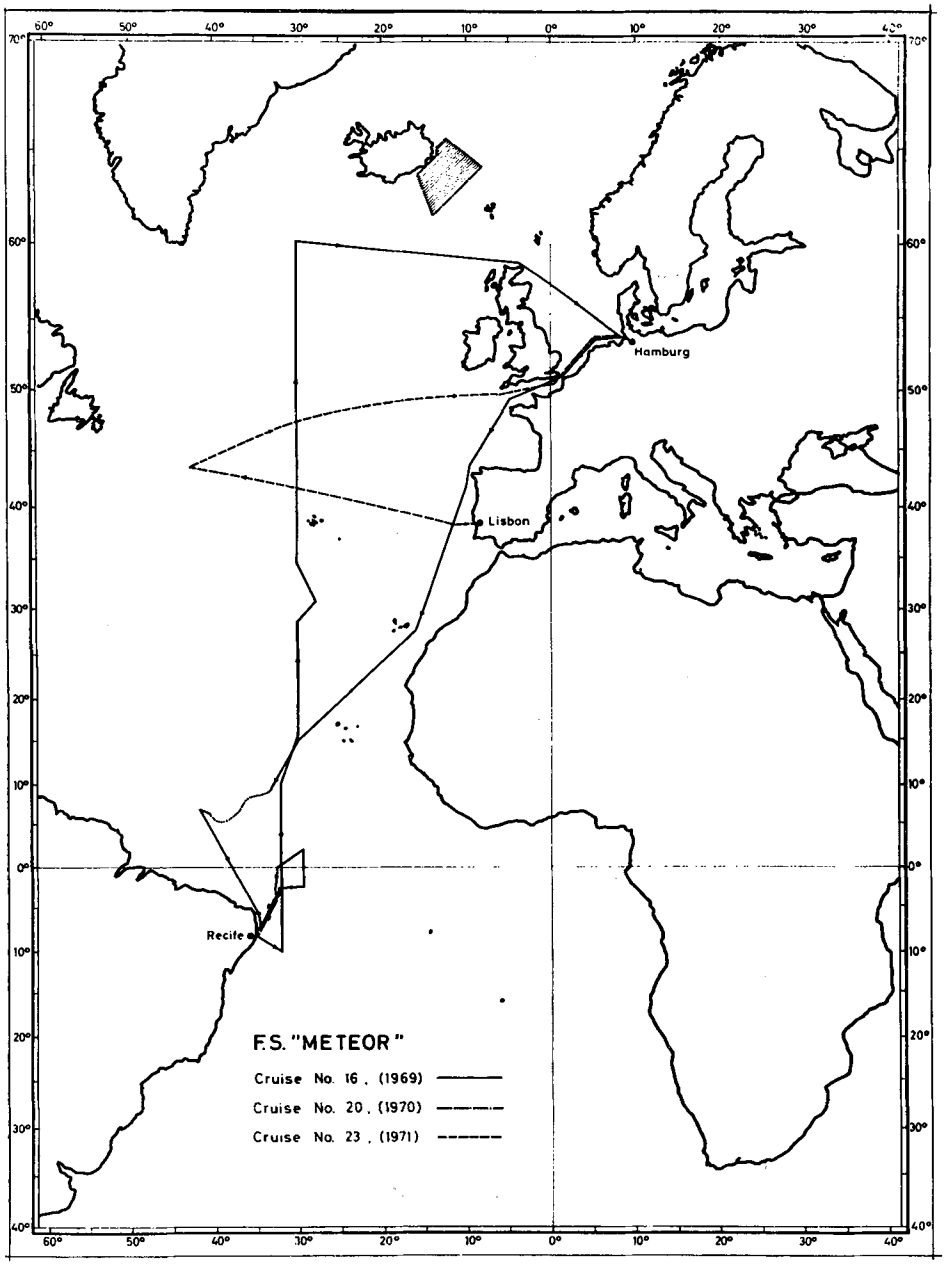

Fig. 1. Map of *Meteor* cruises in 1969, 1970, and 1971.

a Töpler pump was used for the N_2O transfer to the gas chromatograph. After that expedition, the analytical method was improved so that the Töpler pump could be eliminated. The standard deviation of the method with the Töpler pump was $\pm 5\%$ for air samples and $\pm 10\%$ for water samples. The standard deviation of the improved method was found to be $\pm 1.7\%$ for air samples and less than $\pm 3\%$ for water samples.

Table 1. *N_2O data from three expeditions in the North Atlantic Ocean*

	Average mixing ratio in marine air (ppmv N_2O)	Average concentration in sea water (μg N_2O/l)		Average saturation of sea water (% of N_2O eq. conc.)	
		Surface (0–25 m)	Upper layers (0–1 000 m)	Surface (0–25 m)	Upper layers (0–1 000 m)
1969 Expedition (along 30° W)					
(*a*) 0–25° N	0.25 (29)	0.54 (3)	$\geqslant$0.52 (12)	233 (3)	$\geqslant$163 (12)
(*b*) 36–60° N	0.25 (29)	0.50 (4)	$\geqslant$0.45 (12)	141 (4)	$\geqslant$122 (12)
1970 Expedition (Iceland–Faroe ridge)	0.25 (61)	0.45 (20)	0.51 (40)	113 (20)	115 (40)
1971 Expedition (North East Atlantic)	0.27 (34)	0.40 (15)	0.54 (28)	123 (15)	143 (28)

Observations

Fig. 1 shows the map of the 3 cruises. During cruise no. 16—the route is marked on the map by a full line—air samples were taken from January through February 1969 on the voyage from Hamburg to Recife in Brazil and through April 1969 from 10° S to 60° N along 30° W. Sea water samples were taken exclusively during the last part of the cruise.

The N_2O measurements during cruise no. 20 were restricted to that area of the Iceland–Faroe ridge which is hatched. Through June 1970, air and water samples were collected and measured immediately after sampling on board the ship.

During cruise no. 23 in the North East Atlantic ocean—the route is marked on the map by a dashed line—air and water samples were measured through June 1971.

In Table 1 are listed average values which were calculated from the N_2O data we obtained during the 3 cruises. The number of measurements is given in parentheses behind each average value. The first column contains the average mixing ratios in marine air for the last part of the 1969 Expedition and for the entire 1970 and 1971 Expedition. While the 29 N_2O values from 1969 cover a range from 0.18 to 0.33 ppm by volume the scatter of data from the two later expeditions is considerably smaller. In 1970, we found a range from 0.24 to 0.26 ppmv N_2O and in 1971 a range from 0.26 to 0.29 ppmv N_2O.

The second column gives the average N_2O concentrations in sea water in μg N_2O per liter sea water for the surface near water layers down to 25 meters and for the upper 1 000 m. Inspite of considerable temperature changes—from 27°C in the tropical latitudes to 4°C in the area of the Iceland–Faroe ridge —the surface water values change relatively little covering a range from 0.40 to 0.54 μg N_2O per liter sea water.

From this table it is not evident that the surface water value for the region of the Iceland–Faroe ridge is in fact an average of two other average values. The average values from the southwestern slope and the northeastern slope of the ridge of 0.42 and 0.50 μg N_2O per liter sea water are clearly different. The scatter of data indicates the difference to be significant. The reason for this difference is that the water temperatures are changing from about 10°C over the southwestern slope to about 5°C over the northeastern slope of the ridge.

The average values for the upper 1 000 m seem to be in the range of the surface water values. However, as we will see later, the values from the 1969 Expedition must be regarded as lower limits. The values from the 1971 Expedition and—inspite of a better vertical mixing of sea water in the area of the Iceland–Faroe ridge—from the 1970 Expedition, too, seem to indicate a N_2O flux towards the sea's surface. To check this, one should calculate N_2O saturation values taking into account the different water temperatures and the corresponding N_2O mixing ratios in air.

Such values are listed in the third column of Table 1. They can be considered to be correct within $\pm 10\%$. Undersaturation values do not

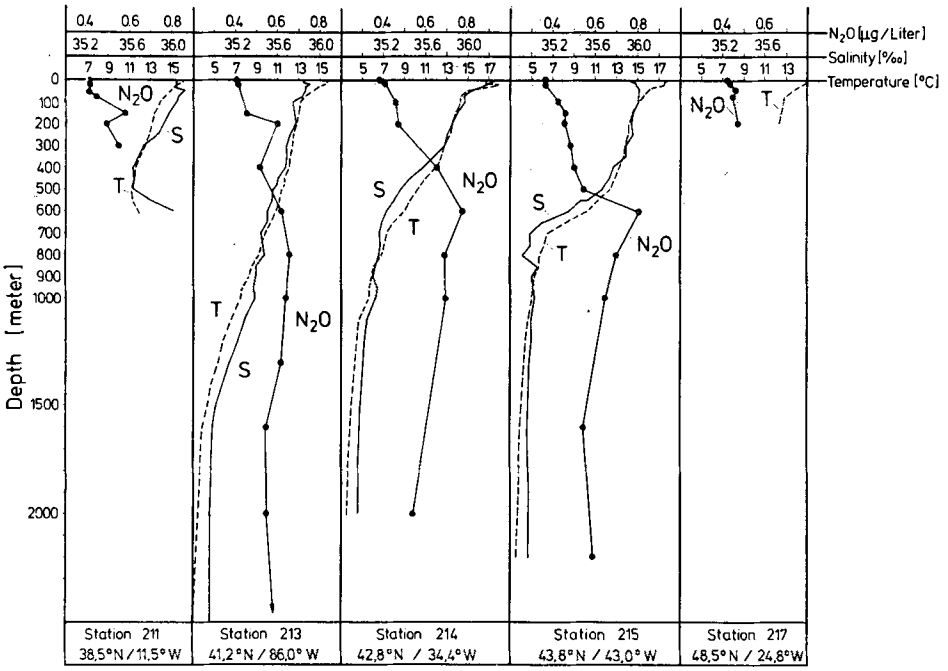

Fig. 2. Vertical profiles of N_2O concentration, salinity, and temperature of sea water at five different stations during the *Meteor*'s cruise no. 23 (1971).

occur. The surface water has supersaturation values up to 230% saturation in tropical latitudes which decrease in a quite systematic way as one travels north.

Except for the data from 1969 which are lower limits, as mentioned above, the supersaturation values for the upper 1 000 meters which are higher than the corresponding surface values show that there must be a N_2O flux towards the surface.

In Fig. 2 are drawn vertical N_2O concentration profiles in sea water together with temperature and salinity profiles from data obtained during the 1971 Expedition at five different stations between the latitudes 38.5° N to 48.5° N. The temperature and salinity curves exhibit the positions of the surface water, the intermediate water, and the deep water layer. At station 211, the increase in temperature and salinity at a depth of 500 m marks the position of a layer of water from the Mediterranean Sea. The more one travels west the more the Mediterranean sea water layer disappears, and at station 213 it is already hard to recognize. The N_2O profiles show two maxima: a small one between 100 and 300 m and a large one between 400 and 1 000 m. These maxima together with the supersaturation values from Table 1 indicate both a N_2O flux towards the surface and the production of N_2O in the sea. N_2O concentrations up to 0.8 μg/liter occur within the large maxima, while the surface water concentrations are around 0.4 μg/liter. At a depth of 1 500 to 2 000 meters there is obviously a minimum, for the N_2O concentration increases slightly again towards greater depths.

During the 1969 Expedition, only a few deep water samples were taken at different stations, mainly from a depth of 50, 100, 500, and 1 000 m. The N_2O profiles from Fig. 2 show (particularly that from station 215) that this is not sufficient to get a proper picture of the situation. For this reason, the average N_2O concentration values obtained from the data of the 1969 Expedition are regarded to be only lower limits.

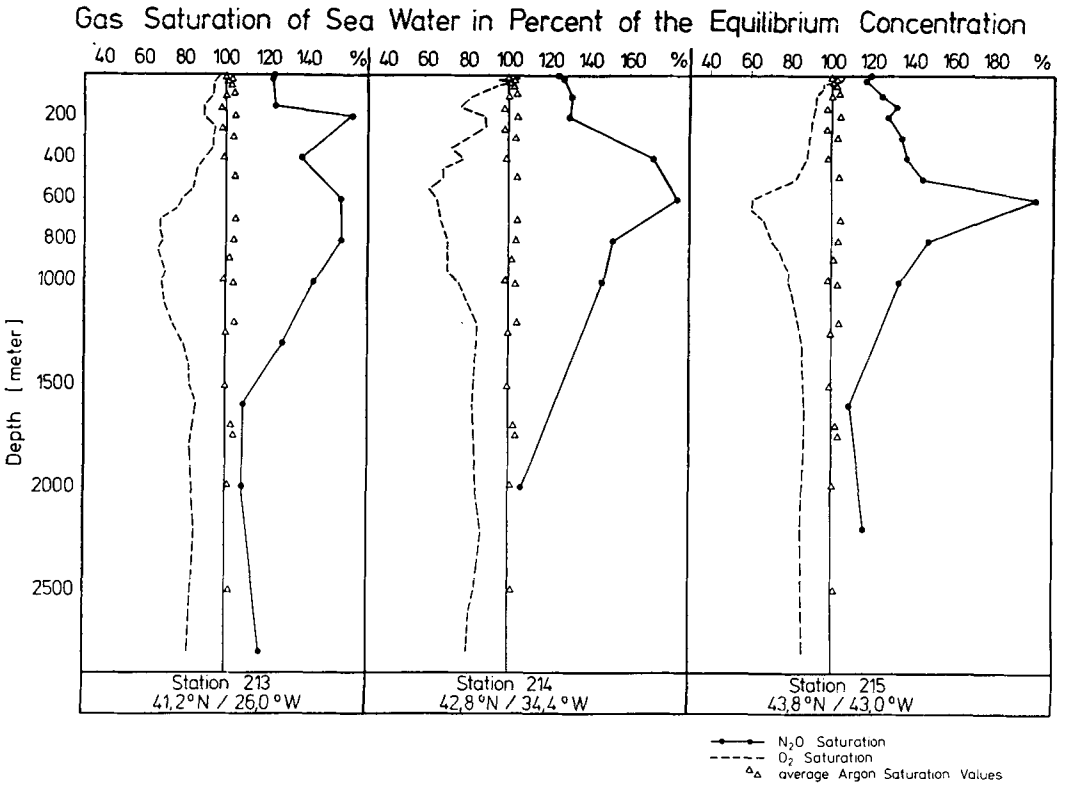

Fig. 3. N_2O and oxygen saturation of sea water as a function of depth at 3 stations during the 1971 Expedition and average argon saturation values from the Woods Hole A-II-20 Expedition (Bieri, 1968) in terms of percent of the equilibrium concentration in sea water.

Discussion

From studies of noble gases in sea water by Bieri et al. (1968) and by Craig & Weiss (1971) it is known that gas supersaturation does not necessarily indicate gas production in the sea. Physical processes such as bubble injection into surface waters and mixing processes of water masses of different temperatures can, as was shown by Kanwisher (1963) and by Bieri (1966), result in gas supersaturation in sea water. Noble gas data from the literature allow us to check the extent that the N_2O supersaturation found may be due to physical processes. The bubble effect will depend on the solubility of the gas in sea water and should decrease with increasing solubility. The solubility of N_2O is about 20 times larger than that for argon. The mixing process of water masses should depend on the second derivative of the variation of solubility with temperature which (in the temperature region of interest) is about 1.5 times that for argon. Thus, the N_2O supersaturation caused by physical processes should be comparable with argon supersaturation percentages.

Fig. 3 shows vertical gas saturation profiles in sea water for N_2O and oxygen calculated from data which were obtained during the 1971 Expedition at the stations 213, 214, and 215 between the latitudes 41° N to 44° N. The oxygen data together with temperature and salinity data were measured by the team of Dr Münnich from Heidelberg. The triangles around the 100% saturation lines mark average argon saturation values calculated from Bieri's 1968 argon data. The argon data were obtained during the Woods Hole A-II-20 Expedition in the tropical North Atlantic ocean (Bieri et al., 1968). Clearer than in Fig. 2 it is demonstrated here to what extent the sea water is supersaturated with N_2O. The 3 diagrams show distinctly that there is at most a small amount of the N_2O supersaturation which may be due to physical processes. Even if the N_2O curves are moved by 10–20% towards the 100% satura-

tion lines, we change no essentials. As a matter of fact we must assume N_2O production in sea water.

As shown by Verhoeven (1952), Wijler & Delwiche (1954), and by Matsubara & Mori (1968) N_2O is produced on land by denitrifying bacteria. Since soil bacteria are also found in water it is not unreasonable to assume a similar mechanism for N_2O production in sea water. There is a series of papers on this topic. In this connection the papers of Cooper (1937), Hutchinson (1944, 1954), Kriss (1949, 1961), Richards & Vaccaro (1956), Richards & Benson (1961), and Brisou & Vargues (1961) should be mentioned. Denitrifying bacteria usually reduce nitrates and nitrites through nitric oxide and nitrous oxide intermediates down to molecular nitrogen. Matsubara & Mori (1968) showed that under certain conditions the reduction step from N_2O to nitrogen can be fully or partly inhibited, so that N_2O is released to the environment. Compared with the N_2O concentrations found (converted: 0.3–0.5 μg N_2O-nitrogen per liter sea water) the nitrate concentration in the North Atlantic sea water, at all depths, is more than sufficient (140–430 μg NO_3^--nitrogen per liter sea water, according to Vaccaro (1965)) for supporting bacterial production of N_2O. Since nitrate is always in excess, N_2O production must be controlled by other parameters.

Fig. 3 shows that minima occur in the oxygen profiles where the N_2O profiles exhibit maxima. Furthermore, there is a fair correlation between the corresponding N_2O and oxygen profiles. This leads to the conclusion that the concentration of free dissolved oxygen is apparently one of the controlling parameters for the N_2O production in sea water, and that in the following way: At depths greater than the photo-synthetic zone the free dissolved oxygen is consumed by respiration and by oxidation of the organic matter, and if there would not be an advective and diffusive renewal of oxygen, it would be completely used up by these processes, as is the case in regions with anoxic conditions. When there is not enough dissolved oxygen available nitrite and nitrate ions supply the reducible substrate for the biochemical oxidation processes. Concurrent with the decrease of free dissolved oxygen, therefore, denitrification increases. Assuming a constant ratio $N_2O:N_2$ in the products of the denitrification process, the N_2O concentration in sea water should increase when the concentration of free dissolved oxygen drops.

Relevant to the question of whether the North Atlantic ocean is a net source of the atmospheric N_2O or not are only the surface water data. Because these data are so critical to the argument, all of the N_2O values obtained during the 3 expeditions from the sea's surface water down to 25 meters are listed in Table 2. The fifth column of this table gives the N_2O concentrations in μg N_2O per liter sea water. It has been noted for the N_2O concentration of the North Atlantic ocean water that the surface water values do not vary so much as one travels south, inspite of a change in water temperature from 4°C in the area of the Iceland–Faroe ridge to 27°C in tropical latitudes. The range of data from 0.37 to 0.65 μg/l N_2O is relatively small. Double measurements show that aboard ship the accuracy of the analytical method used during the 1969 Expedition was within 10–20% (samples 4/5 and 6/7), and was within about 5% with the improved method used during the 1970 and 1971 Expedition (samples 15/16 from the 1970 Expedition and samples 1/2 and 5/6 from the 1971 Expedition). The local variability of the N_2O concentrations found does not show any systematical tendency. Taking into account, however, the variation of the water temperatures by calculating N_2O saturation values for the surface water (tabulated in the last column of Table 2) a very definite trend of increasing N_2O supersaturation of the sea water can be noted as one travels south. This observation already made with the 1969 measurements is well supported by the N_2O data obtained during the 2 later expeditions. The saturation values are less exact than the concentration values, because the N_2O solubility in sea water is not well enough known (Junge et al., 1971). The accuracy of the saturation values from the 1970 and 1971 Expedition can be considered to be better than 15%, that of the 1969 saturation values should be better than 25%. The N_2O supersaturation of sea water found in the area of the Iceland–Faroe ridge is obviously rather small and varying because of the different currents and possible mixing processes of cold and warm water occuring in this region. Although the N_2O supersaturation of sea water found during the 1971 Expedition is, in general, higher, the spread of data is narrower than in 1970. The highest N_2O supersaturation of sea

Table 2. *N_2O in the surface water of the North Atlantic ocean*

Expedition	Sample No.	Position °N	°W	Depth (m)	N_2O concentration (μg/l sea water)	N_2O saturation (% of eq. conc.
No. 16	1	0.6	32	0	0.50	232
(April 1969)	2	10.3	32	25	0.65	293
	3	25.6	30	0	0.46	175
	4	36.6	30	0	0.42	145
	5	36.6	30	0	0.49	169
	6	59.9	30	0	0.50	119
	7	59.9	30	0	0.55	131
No. 20b	1	62.0	12.3	0–2	0.40	107
(June 1970)	2	62.7	11.3	0–2	0.40	106
	3	64.1	8.5	0–2	0.38	104
	4	62.9	11.5	0–2	0.40	106
	5	62.3	13.3	0–2	0.39	104
	6	62.0	14.7	0–2	0.51	135
	7	63.4	15.0	0–2	0.40	109
	8	63.0	19.3	0–2	0.40	109
	9	63.3	18.1	0–2	0.44	119
	10	63.5	14.7	0–2	0.43	118
	11	63.9	13.4	0–2	0.43	115
	12	64.9	8.2	0–2	0.52	113
	13	64.9	8.2	20–30	0.49	110
	14	65.3	9.0	0–2	0.48	108
	15	65.1	11.1	0–2	0.49	112
	16	65.1	11.1	0–2	0.51	116
	17	64.5	13.8	0–2	0.57	123
	18	64.2	14.0	0–2	0.51	125
	19	65.5	11.6	0–2	0.48	110
	20	64.2	13.1	0–2	0.47	115
No. 23c	1	38.5	11.5	0	0.41	125
(June 1971)	2	38.5	11.5	2	0.39	117
	3	38.5	11.5	20	0.42	125
	4[a]	39.9[a]	18.8[a]	0[a]	0.41[a]	126[a]
	5	39.9	18.8	0	0.40	121
	6	39.9	18.8	2	0.39	120
	7	41.2	26.0	0	0.41	124
	8	41.2	26.0	20	0.41	123
	9	42.8	34.4	0	0.38	125
	10	42.8	34.4	15	0.41	128
	11	43.8	43.0	0	0.37	120
	12	43.8	43.0	25	0.37	118
	13	45.8	36.9	0	0.42	130
	14	48.5	24.8	0	0.42	124
	15	48.5	24.8	15	0.43	127

[a] This sample was taken about 1 000 m off the ship.

water was found in 1969 in tropical latitudes. Even if one takes into account the uncertainty of the data, there is no question that the sea water of these latitudes is considerably supersaturated with N_2O. A possible explanation for this can be found in the work of Kriss et al. (1961). This Russian group determined the concentration of heterotrophic micro-organisms in Atlantic sea water from 60° N to the equator along the 30° meridian, and found the highest concentrations of micro-organisms in equatorial sea water with rapid decrease to low concentrations in middle latitudes.

Let us see now how important the oceanic N_2O source may be for the atmospheric N_2O budget as compared with the soil. According

to the stagnant film model (Kanwisher, 1963; Broecker & Peng, 1971) the flux of a gas from the ocean into the atmosphere is given by:

$$\text{FLUX} = \frac{D}{Z}(C^w - P \cdot \alpha(T, S))$$

where C^w is the concentration of the gas in sea water, $\alpha(T, S)$ the solubility of the gas in sea water, D the molecular diffusion rate of the gas in sea water, and Z the film thickness. D/Z is called "piston velocity". P is the partial pressure of the gas in the atmosphere.

In his contribution to the CACGP Symposium on trace gases (April 1973 in Mainz, W.-Germany) W. S. Broecker reported the average N_2O piston velocity for the Atlantic ocean to be 1 200 m per year. Our measurements yield an average N_2O saturation of 180% for the North Atlantic surface water. The area of the North Atlantic ocean is 0.41×10^{14} m². The resulting flux from the surface into the atmosphere is 0.11×10^{14} g N_2O per year. This value can be considered to be correct within one order of magnitude.

The N_2O flux from the land surface into the atmosphere was reported by Schütz et al. (1970) for potting soil to be 0.4×10^{-12} g per cm² and second. N_2O measurements over two years in spring waters from the Taunus mountains (near Wiesbaden, W.-Germany) resulted in an average N_2O flux from the soil into the atmosphere of 0.5×10^{-12} g per cm² and second (Hahn, 1973). When we consider this value as a representative average for the land surface of the northern hemisphere we obtain an overall flux from the land into the atmosphere of 0.16×10^{14} g N_2O per year. Accordingly, the North Atlantic ocean produces nearly as much N_2O per year as the entire land surface in the northern hemisphere does.

In his paper on the exchange of gases between the atmosphere and the sea (1963) Kanwisher plotted relative gas exchange values as a function of latitude in order to demonstrate the effect of the strong non-seasonal westerlies and the greater proportion of water to land in the southern hemisphere on the gas exchange. From this diagram and from Broecker's average piston velocity of 1 200 m per year one can calculate a piston velocity of about 1 700 m per year as a world average. This value corresponds to a film thickness of 28 μ.

When we assume a similar N_2O saturation of the surface water of all oceans, we obtain a global marine net production of 13.5×10^{13} g N_2O per year. With a total mass of 2.0×10^{15} g N_2O in the atmosphere (based on an atmospheric N_2O mixing ratio of 0.25 ppmv) and 2.4×10^{13} g N_2O per year as the global net production of the soil we obtain an atmospheric N_2O life time of 12–13 years. This is in good agreement with the life time of about 10 years given by Schütz et al. (1970) and with the life time of about 12 years given by Broecker (1973).

With the understanding that the assumptions we made and the photochemical destruction rate of about 3.0×10^{13} g N_2O per year (Bates & Hays, 1967; Schütz et al., 1970) are correct, we end up with about 10×10^{13} g N_2O per year which have to be consumed somewhere in the troposphere.

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СЕВЕРНАЯ АТЛАНТИКА КАК ИСТОЧНИК АТМОСФЕРНОГО N_2O

В 1969, 1970 и 1971 гг. во время трех рейсов в Северной Атлантике были предприняты измерения N_2O в морской воде. Пробы морской воды брались начиная от поверхности моря и до глубины 3 000 *м* на нескольких станциях. Концентрации у поверхности в тропическо-субтропических широтах составили в среднем 0,5 *мкг* N_2O на литр морской воды. В районе от 38,5° с. ш. до 48,4° с. ш. было найдено среднее значение 0,4 *мкг* N_2O на литр морской воды. В районе Исландско-Фарерского хребта концентрации у поверхности составили в среднем 0,4 *мкг* на литр морской воды при температуре воды 10°C и 0,5 *мкг* N_2O на литр морской воды при температуре 5°C. Профили вертикальной концентрации часто обнаруживают два максимума: меньший — в слое 100–200 *м* и больший — в слое 400–1 000 *м*, причем концентрация достигает 0,8 *мкг* на литр морской воды. За искюлением нескольких проб, измерения показывают, что морская вода в Северной Атлантике перенасыщена N_2O. Перенасыщения, достигавшие 100 %, были обнаружены в верхних слоях морской воды начиная от тропическо-субтропических широт и до 48,5° с. ш. Далее к северу перенасыщение морской воды N_2O убывает. Для верхних уровней морской воды вплоть до 1 000 *м* были получены следующие средние значения: в тропических широтах перенасыщение составило 66 %, в субтропических широтах — около 47 %, в области от 38° с. ш. до 48,5° с. ш. — около 42 % и в районе Исландско-Фарерского хребта — около 12–20 %. Можно сделать вывод, что Северная Атлантика действует как чистый источник атмосферного N_2O. Повидимому другие океаны обладают той же способностью.