

Reports

Transport and Loss of Nitrous Oxide in Soil Water After Forest Clear-Cutting

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Whole-tree harvesting increased the concentration of nitrous oxide dissolved in soil water by two orders of magnitude over the concentration expected in equilibrium with the atmosphere. In contrast, the nitrous oxide content of soil water in an intact, second-growth forest was close to the expected theoretical value. Nitrous oxide, produced at active sites in the soil, dissolves in soil water and is transported to seeps and streams where it rapidly degasses from the solution and is released into the atmosphere. This loss of nitrous oxide after clear-cutting is not important to the nitrogen economy of the site; however, it may be important to the global atmospheric budget of nitrous oxide. Sources of nitrous oxide may have been overlooked because nitrous oxide emissions can be separated in time and space from the sites of the most intense production of nitrous oxide.

RESEARCH AT THE HUBBARD Brook Experimental Forest (HBEF) in New Hampshire (43°56'N, 71°45'W) has shown that various harvesting practices stimulate the loss of dissolved ionic forms of nitrogen, especially nitrate, in stream water (1). Nitrate loss was attributed to an increase in the activity of autotrophic (2) and possibly heterotrophic (3) nitrifiers. This response is not universal in forests (4), but it is common (5).

We questioned whether the production of NO_3^- after clear-cutting also stimulates the production of gaseous nitrogen compounds (specifically N_2O and N_2) by either nitrification (6) or denitrification (7). Typically, open or closed chambers are used to determine the diffusive flux of N_2O or N_2 from soil directly into the atmosphere (8). However, N_2O is highly soluble in water, and it has been suggested that the transport of N_2O by soil water could be important in some cases (9). We report here that N_2O , produced microbiologically in the soil and dissolved in soil water, eventually emerges in seeps and streams where it degasses into the atmosphere. Emissions of N_2O to the atmosphere may be separated in time and space from sites of N_2O production; consequently, important sources of N_2O may have been overlooked. Water samples were collected from seeps, where saturated flow within the soil emerges naturally to become stream water. Seeps were sampled in a whole-tree harvested watershed and in various 70-year-old, second-growth watersheds at the HBEF. The watersheds range in size from 12 to 43 ha and in elevation from 500 to 800 m. Dominant trees are sugar maple (*Acer saccharum* Marsh.), American beech

(*Fagus grandifolia* Ehrh.), and yellow birch (*Betula alleghaniensis* Britt.), with stands of red spruce (*Picea rubens* Sarg.) and balsam fir [*Abies balsamea* (L.) Mill.] at the highest elevations. Soils are typical frigid Haplorthods (podzols) of the Littleton formation. The HBEF and experimental watersheds are described in detail elsewhere (10).

At each seep two water samples were collected in 50-cm³ glass syringes, fixed in the field, and transported immediately to the laboratory for analysis (11), usually within 5 to 24 hours. Dissolved N_2O in the samples was estimated by multiple equilibration with high-purity helium (12). Nitrous oxide was analyzed by electron-capture gas chromatography (13).

Whole-tree harvesting increased the concentration of N_2O dissolved in soil water by two orders of magnitude over the concen-

Table 1. Decrease in concentration of dissolved N_2O as nitrogen (mean $\pm$ 1 SEM, $n = 2$, unless noted otherwise) between the source and sites immediately downstream from a seep in the cut watershed.

Sample date (1984)	Distance from source (m)	Concentration ($\mu\text{g/liter}$)
23 April	0	1.77 $\pm$ 0.33
	10	0.53 $\pm$ 0.01
13 June	0	2.20 $\pm$ 0.05
	2	1.08 $\pm$ 0.02
9 July	0	7.51 $\pm$ 0.02
	30	2.53 $\pm$ 0.04
21 August	0	56.64 ($n = 1$)
	4	26.12 ($n = 1$)
3 October	0	30.28 ($n = 1$)
	2	9.01 ($n = 1$)
	20	1.11 $\pm$ 0.01

tration expected in equilibrium with the atmosphere (Fig. 1). Figure 1A shows the measured and theoretical values for dissolved N_2O in water from one seep in the whole-tree harvested watershed. The area around this seep was harvested about 6 months before the first sample date. The theoretical value is based on measured water temperature, an assumed atmospheric concentration of 300 parts per billion (14), and the theoretical solubility of N_2O at zero salinity (15). The theoretical value is relatively insensitive to small errors in water temperature or in the assumed atmospheric N_2O content (16). The N_2O content of water from seeps in the uncut, second-growth forest was similar to the expected theoretical value (Fig. 1B).

The content of dissolved N_2O in soil water from cut and uncut areas is controlled by different factors. The solubility of N_2O in water increases as temperature decreases. This physical control is evident in data from the uncut watersheds (Fig. 1, B and C) where the N_2O content in soil water is close to the value predicted on the basis of temperature alone, rising and falling only slightly with seasonal temperature changes. In contrast, the N_2O content of the seep in the cut watershed ranges over two orders of magnitude and peaks in the warmest months (Fig. 1, A and C). The likely sources of this N_2O are nitrification and denitrification, both microbiological processes that are positively correlated with temperature (17).

Nitrous oxide dissolved in soil water rapidly degasses from the solution to the atmosphere (Table 1). It is unlikely that the observed decrease in N_2O is due to biological reduction within the stream (18) since the stream is well aerated and the transit time between the sample pairs in Table 1 is on the order of minutes. Consequently, unlike the loss of ionic forms such as NO_3^- , the loss of N_2O from watersheds cannot be assessed from simple hydrologic and chemical measurements at a downstream outlet.

Elevated levels of N_2O in soil water occur only in the harvested watershed (Table 2). In the harvested watershed area (22.2 ha), the population of seeps is small. Although the variability between seeps and between sample dates is high, dissolved N_2O in soil water tends to be high for all seeps sampled in the harvested watershed and is uniformly low in uncut areas (Table 2).

Approximately 7.3 kg of nitrogen as N_2O (0.33 kg of nitrogen per hectare) were lost from the entire cut watershed by degassing from soil water between April 1984 and October 1985 (19). This increase in export

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Table 2. Variability of dissolved N₂O concentrations as nitrogen (mean $\pm$ 1 SEM, $n = 2$, unless noted otherwise) between different seeps on three dates in the cut and uncut areas.

Sample date	Vegetation type*	Temperature (°C)	Concentration (µg/liter)	
			Theory	Measured
<i>Cut</i>				
21 August 1984	SF	13.9	0.404	0.4 ± 0.01†
	MHW	15.7	0.272	0.4 ± 0.02†
	MHW	22.9	0.215	34.4 (n = 1)
	MHW	20.6	0.232	5.6 ± 0.02
3 October 1984	SF	8.5	0.353	45.2 ± 6.5
	MHW	11.3	0.318	19.8 ± 0.02
6 June 1985	SF	13.2	0.297	495.9 ± 6.4
	MHW	10.6	0.326	34.4 ± 0.5
	MHW	14.3	0.286	164.3 ± 4.7
<i>Uncut</i>				
21 August 1984	MHW	13.0	0.299	0.264 ± 0
	MHW	13.0	0.299	0.274 ± 0.010
	MHW	13.4	0.295	0.376 ± 0.024
3 October 1984	MHW	10.3	0.330	0.343 ± 0.011
	MHW	8.0	0.360	0.398 (n = 1)
	MHW	9.5	0.340	0.382 ± 0.024
6 June 1985	MHW	11.1	0.321	0.226 ± 0.098
	MHW	11.1	0.321	0.142 ± 0.006
	MHW	7.5	0.367	0.480 ± 0.003
	MHW	14.1	0.288	0.231 ± 0.062
	MHW	7.8	0.362	0.212 ± 0.054
	MHW	7.1	0.372	0.432 ± 0.001
	MHW	8.6	0.352	0.494 ± 0.008

*SF, spruce/fir; MHW, mixed hardwoods (maple, beech, and birch); refers to former vegetation on watershed 5. [†]This site was cut less than 1 month before the sample date, so the microbial populations responsible for N₂O production may not have had time to establish. The other sites had been cut for at least 6 months before sampling.

of N₂O by degassing is similar to the measured increase in soil-air diffusion of N₂O from the harvested watershed (20). For comparison, the loss of NO₃⁻ alone from stream water from April 1984 to April 1985 was approximately 729 kg of nitrogen (32.8 kg of nitrogen per hectare) for the same watershed. Consequently, although the concentration of N₂O in soil water increases dramatically after whole-tree harvesting, the mass of nitrogen lost by degassing is probably a small part of the total nitrogen exported from this particular forest after harvesting.

This additional source of N₂O may be important to the global budget of atmospheric N₂O. The existing literature on the relations between landscape disturbance and N₂O emissions to the atmosphere is based on soil-air diffusion measurements. Transport of N₂O by soil water and subsequent degassing have not been quantified for natural disturbances, such as fire or storm damage, or for anthropogenic disturbances, such as other types of forest harvesting practice, urban development, and agricultural practices (especially fertilization and irrigation). The additional export of N₂O is easy to overlook because it may be displaced temporally and spatially from expected sites of

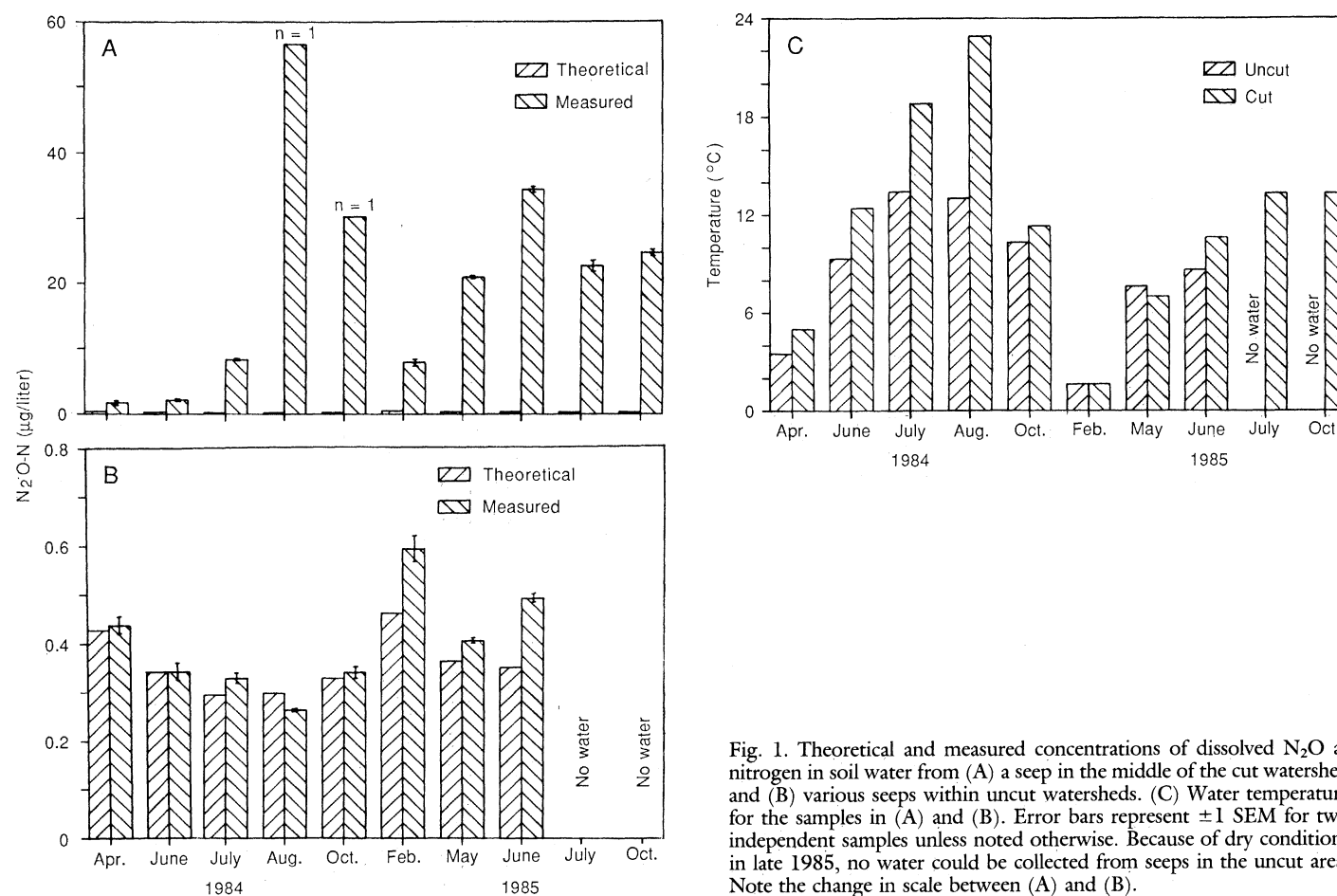


Fig. 1. Theoretical and measured concentrations of dissolved N₂O as nitrogen in soil water from (A) a seep in the middle of the cut watershed and (B) various seeps within uncut watersheds. (C) Water temperature for the samples in (A) and (B). Error bars represent ± 1 SEM for two independent samples unless noted otherwise. Because of dry conditions in late 1985, no water could be collected from seeps in the uncut area. Note the change in scale between (A) and (B).

N₂O production. If the stimulatory effect of soil disturbance on soil-water transport of N₂O is as large at other disturbed sites as it is at HBEF, then the total N₂O emission rate (soil-air diffusion plus soil-water degassing) from disturbed soils may be higher than previously reported (21).

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- Glass syringes (50 cm³) (Becton-Dickinson) were soaked in distilled water and rinsed clean. A plastic, three-way, stopcock (Pharmaseal, American Hospital Supply) was attached to each syringe by a Luc-Lok. The stopcocks and syringes were gas-tight for at least 48 hours. Each syringe was flushed repeatedly and then filled to 20 to 40 cm³ with high-purity helium (Linde, Union Carbide). In the field the helium was discharged under water, and >25 ml of water was drawn into the syringe so as to avoid degassing and inclusion of bubbles. Excess water was discharged so that only 25.0 ml was retained. The sample was fixed in the field with 0.5 ml of 2% (v/v) H₂SO₄ that had been bubbled with helium to remove N₂O. Acid-fixed samples had slightly less N₂O (about 4% after dilution by the H₂SO₄) than unfixed samples. Acidic chemodenitrification of NO₂⁻ or NO₃⁻ would have been expected to increase the N₂O content. At each site two syringes were filled for gas analysis, and two 100-ml polypropylene bottles were filled for pH (Orion, 399 A/F). Water temperature was measured to ±0.1°C with a digital thermometer (Omega, 871) and a thermocouple (Chromel-Alumel, type K).
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- Nitrous oxide in the syringe headspace was measured on a gas chromatograph (Shimadzu, Mini-2) with an electron-capture detector operated at 320°C. The column was 3.1 m of 0.318-cm stainless steel packed with 80- to 100-mesh Porapak Q (Waters Associates) and operated at 70°C with a flow rate of 40 ml/min of a carrier gas mixture of 95% argon and 5% methane (P-5, Linde Gas Products, Union Carbide). Nitrous oxide eluted in about 105 seconds and was quantified against standards (Scott Specialty Gases).
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- All streamflow from these watersheds originates as soil water since there is virtually no overland flow. The total amount of N₂O degassed from soil water was approximated as the averaged volume of seasonal stream water exported from the cut watershed multiplied by the averaged seasonal content of N₂O in soil water, summed over the months of consideration. This is an underestimate because this calculation does not include the volume of water evaporated within the watershed.
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Placers of Cosmic Dust in the Blue Ice Lakes of Greenland

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A concentration process occurring in the melt zone of the Greenland ice cap has produced the richest known deposit of cosmic dust on the surface of the earth. Extraterrestrial particles collected from this region are well preserved and are collectable in large quantities. The collected particles are generally identical to cosmic spheres found on the ocean floor, but a pure glass type was discovered that has not been seen in deep-sea samples. Iron-rich spheres are conspicuously rare in the collected material.

TYPICAL METEORITIC DUST IN THE 0.1- TO 1-mm size range melt during entry into the atmosphere to form "cosmic spheres." The spheres, and the rare particles that enter without melting, are an important resource of extraterrestrial material that is probably a relatively unbiased sampling of the millimeter meteoroids that traverse the inner solar system. Particles of this size produce visual meteors in the night sky and account for the bulk of the 10⁴ tons of extraterrestrial material annually accreted by the earth. The major collection site for cosmic spheres has been deep-sea sediments where spheres are found in moderate concentration because of the exceedingly low accumulation rate of terrestrial particulates. However, even in the deep-sea clays with the highest known concentrations of cosmic dust the spheres larger than 0.5 mm are only 10 parts per billion of the total sediment

mass, and it has not been possible to collect adequate numbers of large spheres and unmelted particles (1). Particles larger than 500 μm are required for certain analysis techniques, such as the measurement of trace elements and isotopic analysis of O, Ti, and the cosmogenic (cosmic ray product) isotopes ²⁶Al, ¹⁰Be, and ⁵³Mn (2).

In addition to deep-sea sediments, it has long been recognized that clean polar ice deposits could be good collection sites for small meteoritic particles (3). Although some spheres have been found in ice, it was not expected that significant numbers of

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