The latest status of nitrogen saturation on Kureha Hill, Toyama, Japan, based on 20-year observations

Miyu Takahashi[1] and Tomonori Kawakami[1]

[1]Toyama Prefectural University

October 4, 2023

Abstract

Excessive anthropogenic nitrogen fixation alters the nitrogen cycle and increases nitrogen deposition, leading to nitrogen saturation, which leads to forest decline and nitrate leakage into stream waters. Kureha Hill in Toyama Prefecture, Japan, is considered to be in nitrogen saturation, since many streams have contained high concentrations of nitrate for more than 20 years. This study verified the latest status of nitrogen saturation by comparing the latest data with 20-year observational data. Results showed that the current nitrate concentration in stream water is lower than that observed 20 years ago. However, the C/N ratio of the soil between 12 and 18, indicates that nitrogen saturation could take place. In addition, the net nitrogen mineralization and net nitrification rate showed no significant change from the past data. Based on the nitrogen leakage to the stream water, Kureha Hill's nitrogen saturation was found to be less intense; however, its potential to produce nitrate has been unchanged for 20 years.

1. Introduction

In addition to the Haber–Bosch process, which made artificial nitrogen fixation possible in 1913, large amounts of nitrogen compounds have been fixed by artificial activities such as the high consumption of fossil fuels. It was estimated that artificial nitrogen fixation had exceeded natural nitrogen fixation by the end of the 20th century (Galloway & Cowling, 2002, Fowler et al., 2013).

Galloway et al. pointed out that the excess nitrogen was retained in groundwater, the soil, or vegetation (Galloway et al., 2008). As a result, nitrogen is no longer a factor limiting the growth of forest ecosystems, and nitrate is leached into stream water (Brown et al., 1988, Galloway et al., 1995). Aber et al. defined this situation as "nitrogen saturation", which takes place when more reactive nitrogen is chronically supplied from the atmosphere to the forest ecosystem than is required by forest ecosystems. Aber et al. proposed four stages of nitrogen saturation according to its progress, as shown in Table 1-1 (Aber et al., 1989).

On the other hand, Stoddard applied the differences in nitrate leakage into stream water to Aber's four stages, as shown in Table 1-2 (Stoddard, 1994).

With regard to nitrogen saturation, many experiments and studies have been done to investigate the response of the ecosystems and nitrate leakage to stream water by adding nitrogen on the forest floor (Molden et al., 1995, Tietema et al., 1997, Wallace et al., 2007, Lundin & Nilsson, 2021, Magill et al., 2004) and on the canopy (Tian et al., 2022), and by cutting vegetation (Likens et al., 1970). Olsson et al. reported the nitrate leaching to stream water when a spruce stand was severely damaged by a windstorm (Olsson et al., 2022). The effects of long-term nitrogen addition on the forest ecosystem and on the nitrate leakage to stream water have been reported recently (Magill et al., 2000, Magill et al., 2004). Not all studies supported the hypotheses advocated by Aber et al. and Stoddard; however, many studies reported phenomena in accordance with the hypotheses (Lovett & Goodale, 2011).

Different from these experimental plots, Kureha Hill, Toyama, Japan seems to have suffered from nitrogen saturation, since many streams on Kureha Hill have high concentrations of nitrate from more than twenty years ago, even though no nitrogen has been added, and no vegetation has been cut (Kawakami et al., 2001). In one of the streams, Hyakumakidani, the nitrate concentration in stream water averaged as high as 160 μmol/L from August 1998 to August 2001 (Honoki et al., 2001), which is higher than that of a Japan-wide stream-monitoring campaign (25.7 μmol/L) (Konohira et al., 2006) and that of the Kanto District of Japan (54 μmol/L) (Nishina et al., 2017). Also, the nitrate concentration in the stream water exceeded the deposition of nitrogen by precipitation (Kawakami et al., 2001). In addition, the nitrate concentration seems to be regulated only by the flow rate of the stream, and uptake by vegetation resulted in no observable impact. Therefore, according to Stoddard's definition (Stoddard, 1994), the nitrogen saturation in Kureha Hill is considered to be Stage 3.

In addition, the stream water of Hyakumakidani is acidified to a pH level of 5.2 (Honoki et al., 2001), and high concentrations of aluminum are being leached (Honoki et al., 2001).

In this study, the 20-year time trends of nitrate concentration and other water qualities, as well as net nitrogen mineralization and the net nitrification of the soil on the nitrogen-saturated watershed and stream water, are discussed.

Site description

Kureha Hill is located in Toyama Prefecture, Japan (36°41' N, 137°09' E). Figure 1-1 shows the location of Kureha Hill.

Kureha Hill is a small mountain formed by a fault, and it stretches for 2 km from north to south. Its highest altitude is 145 m. It overlays alternating layers of sand and mud deposited by Quaternary Lake (Fujii & Yamamoto, 1979). Eighty percent of the hill is covered by deciduous trees, primarily 60-year-old Konara (*Quercus serrata*). The rest of Kureha Hill is covered with bamboo (*Phyllostachys pubescens*) and afforested 70-year-old coniferous Sugi (*Cryptomeria japonica*). The average annual precipitation at Toyama City's weather station, located 1 km from Kureha Hill, is 2388 mm (Japanese Meteorological Agency). No agricultural areas exist within these watersheds. There are many small streams, and the characteristics of the stream water include low pH, low acid-neutralizing capacity (ANC), and low concentrations of base cations. The nitrate concentrations are high throughout the year (Honoki et al., 2001). The Hyakumakidani stream is one of the streams on Kureha Hill whose water quality has been intensively investigated for more than 20 years.

In the Hyakumakidani watershed, nitrate that exceeds the nitrogen supply from the atmosphere by rainfall leaches into the stream water. Figure 1-2 shows the nitrogen budget in the Hyakumakidani watershed from 1999 to 2002 (Honoki et al., 2001). The entire area of Kureha Hill is considered to be nitrogen saturated, since many streams contain high concentrations of nitrate. However, a recent report indicated that the concentration of nitrate was not as high as before (Takahashi & Kawakami, 2023).

2. Methods

2-1. Periodic sampling and analysis of stream water on Kureha Hill

Basically, stream water sampling was conducted biweekly at the Hyakumakidani stream. In addition to the Hyakumakidani stream, stream water was sampled at four other locations (Kanaya, Teramachi, Anyobo, and Chayamachi) to compare the data with that from more than 20 years ago. Figure 2-1 shows the sampling locations on Kureha Hill.

2-2. Measurement of the flow of Hyakumakidani on Kureha Hill

The flow rate was measured in the Hyakumakidani stream. A triangle notch weir was installed in the Hyakumakidani stream, and the upstream water level was measured every 10 minutes, from which the flow rate was determined.

2-3. Nitrogen budget in the Hyakumakidani watershed

Input was calculated from the precipitation observed at the meteorological station in Toyama City 5 km away from Hyakumakidani and the total nitrogen concentration in the rainwater collected by a bulk sampler. Output was calculated from the flow rate of the Hyakumakidani stream observed every 10 minutes and the nitrate concentration estimated from the flow rate.

2-4. Soil analysis

Soil samples taken from depths of 5–10 cm at the three plots within the Hyakumakidani watershed were mixed to produce composite samples for analyzing carbon and nitrogen contents. After taking a moderate amount of air-dried soil and measuring its mass, carbon and nitrogen were measured using a CN coder to determine the C/N ratio of the soil.

2-5. Measuring thenet nitrification rate and net nitrogen mineralization rate

The net nitrification rate and net nitrogen mineralization rate were measured at 3, 15, and 30, using the composite samples according to Honoki et al. (Honoki et al., 2001). The increase in nitrate before and after incubation was defined as *net nitrification* , and the increase in nitrate plus the increase in ammonium was defined as*net nitrogen mineralization* .

3. Results and discussion

3-1. Pattern of nitrate leakage

Figure 3-1 shows the change in nitrate concentration in the Hyakumakidani stream over approximately 24 years, from July 1998 to December 2022. Nitrate concentrations fluctuated greatly according to the stream's water flow (Kawakami et al., 2001).

Figure 3-2(a) shows the nitrate concentration change by flow rate in the Hyakumakidani stream from August 1998 to July 1999. The nitrate concentration increased with the increase in flow rate. The phenomenon, in which the nitrate concentration increases with an increase in the flow rate, has been reported in many rivers (Baures et al., 2013). Sase et al., Kamisako et al., Mitchell et al., and Shibata et al. reported that the nitrate concentration increased with an increase in the flow rate at a site in Japan where nitrogen saturation is suspected (Sase et al., 2019, Kamisako et al., 2008, Mitchell et al., 1997, Shibata et al., 2001). However, the unique characteristic of the Hyakumakidani stream is that a linear relationship was obtained when the logarithm of the flow rate was taken, as shown in Figure 3-2(b). Linearity was maintained with a 100-fold increase in the flow rate from approximately 0.1 L/s to 10 L/s. This indicates that the nitrate concentration seemed to be determined by the flow rate and was not affected by the season. To clarify the independence of the nitrate concentration from the season, nitrate concentrations in the growing season and the dormant season were considered separately.

Figure 3-3 (a) shows the tree growth as measured by dendrometers attached to 6 deciduous stands and 6 coniferous stands randomly selected on the Hyakumakidani watershed. The average relative trunk diameters of both the coniferous and deciduous stands against those of April 2000 are indicated in the figure. Since both coniferous and deciduous stands grew between May and August, the plots in Figure 3-2 (b) were separately plotted again in Figure 3-3 (b) for the growing season (May to August) and for the dormant season (September to April). There was no difference in nitrate concentration between the growing season and the dormant season. This meant that the Hyakumakidani stream was in Stage 3 of nitrogen saturation, according Stoddard's definition, in which there are no seasonal changes in nitrate concentration, and the nitrate concentration exceeds that of precipitation (Stoddard, 1994).

Mitchell et al. reported that Japanese streams containing high concentrations of nitrate generally had less fluctuation in nitrate concentration. They pointed out the Japanese meteorological features, in which, during the growing season, substantial precipitation combined with high temperature enhances nitrogen mineralization and nitrate leakage to stream water, causing less nitrate fluctuation (Mitchell et al., 1997). On the contrary, Kureha Hill is located on the Japan Sea side of Japan, which receives a large amount of snowfall in the winter. Different from the watersheds on the Pacific Ocean side of Japan investigated by Mitchell et al., the flow rate in the dormant season is generally higher than that in the growing season. Therefore, there should be other reasons for the low fluctuation in nitrate concentration. One reason may be less utilization of nitrogen than in the nitrogen cycle of the forest ecosystem of Kureha Hill. Makino et al. conducted a statistical analysis of the nitrate concentration of stream water in the Kinki District to compare the nitrate concentrations in the regions on the Japan Sea side and on the Pacific Ocean side. They concluded that the lower nitrate concentration on Japan Sea side than that on Pacific Ocean side is caused by the flushing of nitrogen in the soil and dilution by heavy rain and snow on Japan Sea side (Makino et al., 2021, Makino et al., 2023). Since Kureha Hill is located on the Japan Sea side and receives a large amount of snow, the flushing of nitrogen and dilution could take place. However, the nitrate concentrations in the streams are quite high.

Figure 3-4 shows the relationship between the flow rate and nitrate concentration in the Hyakumakidani stream in 2019. The trend for nitrate concentrations to increase as the flow rate increased was the same; however, a linear relationship was observed between the nitrate concentration and flow rate without a logarithm in 2019.

Figure 3-5 shows the change in the pattern of nitrate leakage according to the flow rate. When the flow rate was less than 7 L/sec., the nitrate concentration in 1999 exceeded that in 2019. The nitrate concentrations in 1999 and 2019, at an annual average flow rate of 1.4 L/sec in 2009, were 106 μeq/L and 69.4 μeq/L, respectively. This indicates that the nitrate concentration decreased to 65.5% in 2019, as compared with the average flow rate in 2009.

Since the nitrate concentrations were a function of the flow rate, and seasonal variations were not found in either 1999 or 2019, the nitrate concentrations in 1999 and 2019 were estimated from the flow rate, and the nitrate leaching was calculated.

The nitrate concentration in each year was estimated by using the relationship between the flow rate and the nitrate concentration in 1999 and 2019, as shown in Figures 3-2 and 3-4, respectively. In order to compare the nitrate concentrations with the same flow rate pattern, the flow rate in 2009 was used as an example, since the precipitation in 2009 was 2224 mm which was equivalent (93%) to the average precipitation (Japanese Meteorological Agency). Figure 3-6 shows the flow rate in 2009 and the estimated nitrate concentrations by 1999 and 2019. According to the equation, the relationships were estimated to be 2175 mol/ha/year and 1887 mol/ha/year in 1998 and 2019, respectively. The nitrate leakage was reduced to 87% over the 20-year period, which was not as low as the reduction rate of 65.5% when it was compared at the average flow rate. This is because the nitrate ion concentration given by the 2019 model formula at high flow rates (>7 L/sec) exceeded the 1999 model formula, and the 2019 model's high flow rate and high nitrate concentration gave a large quantity of nitrate lodgings. In 2009, a high flow rate of more than 7 L/sec. took place several times in July and November, which caused high loading of nitrate.

Figure 3-7 compares the nitrate concentrations at each site on Kureha Hill between 2005–2006 and 2019. Flow rates at the Hyakumakidani stream were used for comparison, since flow rates were measured only at the Hyakumakidani stream. For 2019, the nitrate concentrations at each site are shown when the flow rates in the Hyakumakidani stream were 0.12, 1.65, and 6.90 L/sec. For 2005–2006, nitrate concentrations at these flow rates were estimated from the equations of the relationship between the flow rate and nitrate concentration in 2005–2006. As mentioned previously, in 2009, the average annual flow rate in the Hyakumakidani stream was 1.4 L/sec. Therefore, the flow rate of 0.12 L/sec. corresponds to a low flow rate, 1.65 L/sec. corresponds to the average flow rate, and 6.90 L/sec. corresponds to a high flow rate. As a whole, nitrate runoff concentrations in 2019 were less than those in 2005–2006, when compared at the same flow rate. It is apparent that the nitrogen input to the catchment by deposition or addition is one of the factors that regulates the nitrate concentration in the stream water (Tietema et al., 1997, Nishina et al., 2017, Ohrui & Mitchell, 1997, Dise & Wright, 1995). Some reports demonstrated a decrease in the nitrate concentration of stream water with a decrease in atmospheric nitrogen deposition due to recent air pollution control measurements (Eshleman et al., 2013, Chiwa, 2021, Gilliam et al., 2019, Baba et al., 2020).

3-2. Nitrogen budget

Figure 3-8 shows the nitrogen budget in the Hyakumakidani watershed from 2019 to 2021 in addition to from 1999 to 2002. The average inputs and outputs for 2019–2021 were 1046 eq/ha/year and 1097 eq/ha/year, respectively. In contrast, the average inputs and outputs for 1999–2002 were 1598 eq/ha/year and 2317 eq/ha/year, respectively. Compared to 1999–2002, both input and output decreased significantly from 2019 to 2021. Accordingly, the decrease in output could be attributed to the decrease in input. However, when taking consideration of other streams than the Hyakumakidani stream (Fig 3-7), the Kanaya stream showed the same nitrate concentration while the Anyobo stream showed a drastic decrease. There must be other factors regulating nitrate concentration such as nitrogen pool in the soils (You et al. 2001). In 1999–2002, output exceeded input every year. However, output became smaller than input in 2020 and 2021. The nitrogen saturation is considered to be mitigated or slowed in its progress.

3-3. pH

Figure 3-9 shows the change in pH in the Hyakumakidani stream over 24 years, from July 1998 to December 2022. No significant changes have been observed over the past 24 years, with values around 5.2 and a minimum value of 4.4. The adverse effects of low pH on fish reproduction and mortality have been well documented, since acid rain caused the acidification of river and lake waters in Europe and North America (Wright & Snekvik, 1978, Baker, 1990, Wignington et al., 1990), and the threshold level of pH could be 5 for acid-sensitive fish, such as trout and salmon— especially their fry (Beamish et al., 1975, Farmer et al., 1980). Since all of the streams on Kureha Hill are small-scale streams with steep slopes, no fish live there; however, the water quality of the Hyakumakidani stream is crucial for fish.

Figure 3-10 shows the change in pH with flow rate in the Hyakumakidani stream, showing that pH decreased as flow rate increased. Taking the H^+ concentration into consideration, when the pH value is reduced from 5.2 to 4.4, which corresponds with the average value and the lowest value, H^+ is increased by 34 μeq/L. On the contrary, the nitrate concentration varied more than 150 μeq/L, indicating that a neutralizing process took place to suppress change in the H^+ concentration.

Figure 3-11 shows the change in the acid-neutralizing capacity (ANC) of the stream water against the flow rate. With an increase in the flow rate, the ANC decreased, indicating that the ANC was consumed to neutralize H^+associated with nitrate. However, the ANC value in the Hyakumakidani stream was originally low due to the geology and was not sufficient to neutralize the entire increase in H^+ associated with nitrate.

Figures 3-12 and 3-13 show the relationship between the sum of cations and chloride concentrations and the flow rate, respectively. No relationship was found, indicating that these ions did not contribute to neutralization.

Figure 3-14 shows the relationship between sulfate concentration and flow rate. The sulfate concentration comparatively decreased according to the increase in the flow rate. The decrease in the sulfate concentration combined with the decrease in ANC is the reason that the H^+ concentration did not increase due to the increased nitrate concentration in the high flow rate.

Figure 3-15 illustrates the inverse relationships between nitrate concentrations and sulfate concentrations for the years 1998–2007, 2008–2017, and 2018–2022. The decrease in sulfate concentration prevents acidification due to the increasing nitrate concentration; however, the sulfate concentration at the specific nitrate concentration gradually decreased over time, which means that sulfate's neutralizing capacity gradually decreased.

3-4. Al concentration

Figure 3-16 shows the relationship between nitrate concentrations and aluminum concentrations. Over the past 24 years, aluminum concentrations increased as nitrate concentrations increased. The low pH caused by the high nitrate concentration in the high flow rate brought high concentrations of aluminum. Since fish mortality is reported with an aluminum concentration of 11 μeq/L (Baker & Schofield, 1982), the aluminum concentration of the Hyakumakidani stream sometimes exceeded the fish mortality threshold level.

3-5. Nitrogen content and carbon–nitrogen (C/N) ratio of the soil

Table 3-1 shows the nitrogen content and C/N ratio of the soil in 2000, 2002, 2019, and 2023. From 2000 to 2023, the nitrogen content in the soil fluctuated greatly. This could be due to spatial inhomogeneity. The C/N ratio also fluctuated between 12 and 18; however, all data showed C/N ratios below 25, which is one indicator of nitrogen saturation (Emmett et al., 1998). Accordingly, the nitrogen saturation of the Hyakumakidani watershed is still considered to have been taking place for more than20 years.

3-6. Net nitrification and net nitrogen mineralization rates

Figure 3-17 shows the net nitrification and the net nitrogen mineralization rates in the soils of the Hyakumakidani watershed at each temperature in 2002, 2019, and 2023. In 2019, the net nitrogen mineralization rate at 30°C showed a particularly high value, and it was much higher than the net nitrification rate. Comparing the net nitrification rate, no significant changes were observed between 2002 and 2023. The potential to produce nitrate has been unchanged for 20 years. Similar values in net nitrification and net nitrogen mineralization rates, which were characteristic of Stage-3 nitrogen saturation according to Aber's prediction, reflect that the Hyakumakidani watershed is considered to remain in Stage-3 nitrogen saturation.

4. Conclusion

Nitrate leakage to some of the streams on Kureha Hill, Toyama, Japan tended to decrease in the 24 years from 1998 to 2022. The reduction in nitrate leakage could be attributed to the reduction in nitrogen deposition. However, since the rate of reduction in nitrate leakage varied across rivers, there must be other factors regulating nitrate concentrations, such as nitrogen pools in the soil. The Hyakumakidani stream, one of the streams on Kureha Hill, had an acidic pH of 5.2 due to the leached nitrate. Despite the recent decline in nitrate concentration, the pH level has been unchanged. This may be due to decreased concentrations of sulfate and ANC.

The C/N ratio of the soil in the Hyakumakidani watershed ranged from 12 to 18 from 2000 to 2023, which is lower than the nitrogen saturation threshold of 25. Net nitrification rate and net mineralization rate showed no significant changes between 2002 and 2023. The potential to produce nitrate has been unchanged for 20 years. Similar values in net nitrification and net nitrogen mineralization rates, which are characteristic of Stage-3 nitrogen saturation according to Aber's prediction, reflect that the Hyakumakidani watershed remains in Stage-3 nitrogen saturation.

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Stage	Description
Stage 0	Tree growth and litter decomposition are limited by nitrogen. No nitrate leaches into stream water because the fo
Stage 1	When the nitrogen supply increases chronically, the foliar nitrogen content increases temporarily. Although the fi
Stage 2	The nitrogen supply exceeds the forest's absorption capacity, and forest ecosystems are limited by water and phos
Stage 3	Nitrate leakage into stream water and N_2O emissions into the atmosphere further increase, and the forest declines

Table 1-1 Aber et al., proposed four stages of nitrogen saturation, depending on the forest ecosystem response, such as foliar biomass and foliar nitrogen concentration.

Stage	Description
Stage 0	The nitrogen cycle is dominated by forest and microbial uptake. Nitrate concentrations are very low throughout t
Stage 1	The nitrogen cycle is still dominated by forest and microbial uptake. Although little nitrate leaches during the gr
Stage 2	The nitrogen cycle is dominated by nitrate leakage and denitrification instead of forest and microbial uptake. The
Stage 3	The nitrogen retention capacity of the forest decreases considerably, and nitrate leakage exceeds nitrogen depositi

Table 1-2 Stoddard, following Aber et al., defined four stages of nitrogen saturation according to the differences in nitrate leakage into stream water.

Year	2000	2002	2019	2023
Nitrogen content (g-N/100 g dry soil)	0.11	0.55	0.58	0.34
C/N	13	18	12	17

Table 3-1 The nitrogen content and C/N ratios of the soil of the Hyakumakidani watershed in 2000, 2002, 2019, and 2023. From 2000 to 2023, the nitrogen content in the soil fluctuated greatly. The C/N ratio also fluctuated between 12 and 18.

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