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学 位 論 文 題 名

Factors controlling temporal and spatial patterns of nitrogen export in Shibetsu watershed, eastern Hokkaido, Japan

（道東標津川流域における窒素流出の時空間変動の支配因子）

学位論文内容の要旨

Humans continue to transform the global nitrogen (N) cycle at a record pace, reflecting an increased combustion of fossil fuels, growing demand for N in agriculture and industry, and pervasive inefficiencies in its use (Galloway et al., 2005). As a result, the amount of reactive N that enters the biosphere each year worldwide has roughly doubled since pre-industrial times (Galloway et al., 1995). Much anthropogenic N is lost to air, water, and land to cause a cascade of environmental and human health problems. For example, riverine N exports, which account for 20-60% of N inputs at watershed scale (Van Breemen et al., 2002), have led to the occurrence of eutrophication in the water body. With the success in control of point source pollution, the non-point source pollution, especially the agricultural N pollution, has become major cause for water pollution. To mitigate the water pollution associated with agricultural N losses, investigate the transport processes and export mechanisms of N from agricultural areas to aquatic systems is crucial. However, the processes and mechanisms of N export are complicated due to a variety of factors, such as land use, topography, hydrological characteristics, soil types, vegetation types, N source, and climatic variables, all of which contribute to different spatial and temporal patterns of N export. Therefore, the objective of this study was to investigate the N export in watershed by using a combined field study and model simulation, and to determine the factors control on N export at heterogeneous spatial and temporal scale.

1. The impact of hydrological process on river N export was investigated in stream water during four storm events in 2003 in the Shibetsu watershed. The results showed that particulate N quickly responded to rainfall and peaked before discharge peak during all storm events; while dissolved N peaked just following the peaks of the shallow groundwater table. The M (V) curve, defined as nutrient mass distribution vs. the volume of discharge, showed a “first flush” for all N components. However, particulate N contributed over 80% of fluxes during the first 50% of the discharge; while dissolved N only released 50% of fluxes during the same stage. Thus the significant flush of particulate N was likely to derive from soil erosion and relate to surface runoff; while the slowly export of dissolved N might originate from the near-surface soil layer associated with the rising shallow ground water table, and then flushed with subsurface runoff.

2. Shibetsu watershed had a coupled land use and topography characteristics which are described as agriculture area with flat topography and forest area with steep slope. To investigate the effect of the coupled characteristics on N export, this study was investigated in three adjacent headwater streams (agriculture-dominated watershed: AW; forest-dominated watershed: FW; and the mixed agriculture-forested watersheds: AFW) in Shibetsu watershed during 2003-2005. The monitoring was conducted from monthly baseflow, over 20 rainfall events and three snowmelt seasons for each watershed. The results showed that higher NO_3^--N concentrations were observed in the agriculture watershed, lower in the forest watershed, and medium in the mixed watershed. A negative exponential relationship (R^2=0.33, P<0.01) was found between the relief ratio (the difference between maximum and minimum elevation of a watershed divided by its maximum length of the river or stream) of the three watersheds and the normalized time by peak discharge time when NO_3^--N peaked. We observed the NO_3^--N concentrations peaked before the peak of discharge in the FW for all hydrological events, regardless of the difference in hydrological characteristics. The quick release of NO_3^--N was attributed to “flushing mechanism”, which was driven by fast response of subsurface flow due to the macropores in forest soil and the steep slope. The AW showed a consistent “prolonged flush” of NO_3^--N, where NO_3^--N concentrations peaked after the peak of discharge, which might attribute to the slow occurrence of subsurface flow because of

the flat slope and low hydraulic conductivities in the pasture.

3. Nitrate-N export associated with water source and water connectivity of hillslope was studied using a combination of field study in a hillslope of upland-riparian zones and watershed scale monitoring within a 36.6 km^2 headwater stream AFW in Shibetsu watershed from July to November 2009. The measurements included monthly water/soil sampling and 15 min interval groundwater table recording at hillslope and monthly water sampling during base flow and hourly water sampling during rainfall events at the outlet of AFW. The results illustrated that during baseflow, low NO_3^--N concentrations (0.06-0.77 mg L^{-1}) were found in upland, riparian forest soil water, and shallow groundwater, but extremely high concentrations (4.44 mg L^{-1}) were shown in spring water (deep groundwater). Deep groundwater was the main source for NO_3^--N export during baseflow. End-member mixing analysis (EMMA) indicated that upper forest stream water, soil water and spring water were the main water sources of streamflow during rainfall events. Soil water was the largest changing part among rainfall events depending on water connectivity of upland-riparian zones. When water connection was only established from riparian zones to stream at the early stage, soil water accounted for small part of streamflow and exported NO_3^--N mainly within riparian zones. When water connectivity was well built from upland to stream, soil water increased the contribution to streamflow sharply and became the main source to exported NO_3^--N. However, large amount of soil water with low NO_3^--N concentrations at the late connection stage suggested the N source was limited, which might be regulated by denitrification process when the water connectivity was well connected. The NO_3^--N export was changed from water source control to N source control.

4. Upscaling the temporal and spatial patterns of N export was performed by SWAT (Soil and Water Assessment Tool) model using 90 m digital elevation model (DEM), 1:50000 soil map, 1:25000 land use map, weather data (from 5 weather stations), and management data (fertilization, grazing and harvest). It is challenge to apply SWAT directly in Shibetsu watershed because of the watershed with external contribution (EXT). In this study, the water flux (1.38 mm d^{-1}) of EXT was roughly estimated by water balance equation using 30 yrs measured data; the NO_3^--N loading of EXT was calculated as 180 kg d^{-1}, based on the NO_3^--N concentration in deep groundwater. The EXT was assigned as a point source discharge and NO_3^--N loading in SWAT to access the model calibration and validation. The simulation of daily streamflow produced satisfactory results with R^2 values of 0.65 and 0.66; and monthly simulation of NO_3^--N achieved a Nash coefficient (Nash and Sutcliffe, 1970, indicates how well the plot of the observed values versus the simulated values fits the 1:1 line) of 0.89 and 0.72 during calibration (2004-2006) and validation period (2007-2008). The result indicated that the application of SWAT in Shibetsu watershed by adding EXT as assumed point source discharge and NO_3^--N loading was successful. In addition, the model upscaled the NO_3^--N export at 30 yrs-time scale (1980-2009). The simulation showed the integrated effect of land use, topography and denitrification process related to soil type regulated the spatial patterns of NO_3^--N export; the hydrological and biogeochemical processes control on NO_3^--N export at temporal scale.

In conclusion, the N export showed significant difference at spatio-temporal scale, which was controlled by different factors, such as hydrological, biogeochemical, land use, topography, water source, and agriculture managements. The best management practices should be taken to effectively control the local agricultural N pollution, based on a well understanding of the N export mechanisms and their factors.

学位論文審査の要旨

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（道東標津川流域における窒素流出の時空間変動の支配因子）

本論文は英文 192 頁, 図 57, 表 27, 9 章からなり, 参考論文 1 編が付されている.

産業革命以降, 過去 100 年以上にわたり, 人類は, 主に農地開発, 農業の化学肥料, 家畜生産, 化石燃料燃焼の増大により, 世界の窒素循環をおよそ二倍にした。このことは環境を巡る窒素量を増加させ多くの環境問題を引き起こしている。水圏への窒素負荷は, 窒素投入量の 20-60%を占める。点源からの負荷が改善される中で、農業由来の面源からの負荷が水質汚染の原因となっている。窒素損失と関連した水質汚染を軽減するためには, 土壌中の窒素の移動と河川への流出に関する研究が必要である。しかしこれらは, 土地利用, 地形, 土壌, 植生, 気候, 水文などが複雑に作用し, 大きな時空間変動をもたらす。本研究の目的は、北海道標津川流域を対象に現地調査とモデルによるシミュレーションを用いて流域の窒素流出に関わる支配要因を決定することである。

河川への窒素流出への水文過程の影響を 2003 年の 4 回の降雨イベントで解析した。粒子状窒素は素早く雨量に応答し, すべての降雨イベントで水量ピークの前に窒素濃度のピークが出現した。一方, 溶存態窒素は浅い地下水位の最高値の後で濃度ピークが観測された。M(V)カーブ（流出量に対する栄養塩流出量の相対分布）は、すべての形態の窒素が水より早く流出することを示した。ただし粒子状窒素の流出は, 水量が 50%流出するときに 80%以上となったのに対し, 溶存態窒素はほぼ水量と同じ 50%の流出を示した。これらのことは, 粒子状窒素の河川水に先行する流出が表面流去水に関連する土壌侵食に由来しており, 溶存態窒素の総体的にゆっくりとした流出は, 上昇する地下水面に関連し流出源は土壌中である可能性を示唆する。

標津川流域は, 平坦な農耕地と急峻な森林という土地利用と地形の特徴をもつ。土地利用と地形が窒素流出におよぼす影響を、近接する 3 つの小流域（AW：農耕地流域、FW：森林流域、AFW：農耕地-森林複合流域）の河川での 2003-2005 年の観測データを用いて解析した。その結果, 河川水の硝酸態窒素濃度は, AW で最も高く, AFW, FW の順に低くなった。起伏比（流域の最大と最小標高差と河川長の比）と相対的な硝酸態窒素濃度のピーク出現時間の間には, 負の指数関係（$R^2=0.33$, $p<0.01$）が認められた。FW では, 降雨量に関わらず全ての降雨イベントで, 硝酸態窒素濃度ピークは流量のピークの前に出現した。この早い流出は、森林土壌で粗孔隙が多いこと, 急峻な地形によると

推察した。AW では，常に硝酸態窒素濃度ピークは流量のピークの後に出現し，牧草地の平坦な地形，低い透水性によって水平流の発生が遅れるとことによると推察した。

AFW の草地−河畔林での，浅層地下水位と河川水位，土壌溶液，地下水，河川水中の窒素濃度の観測データを用いて，流出源と土壌中の水の連結性が窒素流出におよぼす影響を解析した。その結果、草地、河畔林の土壌溶液、浅層地下水の硝酸態窒素濃度は低かったが（0.06−0.77 mg L^{-1}）、湧水（深層地下水）の硝酸態窒素濃度は極めて高かった（4.44 mg L^{-1}）。エンドメンバー法での解析によれば，上流の森林河川水，土壌溶液，湧水が，降雨時の硝酸態窒素の流出源であることが示された。土壌溶液の割合が最も高かったが，草地−河畔林の水の連結性に影響され，降雨イベント間でその比率は異なった。草地−河畔林の連結性が乏しく河畔林と河川のみに連結性が見られるイベント初期には，河川水の土壌溶液の占める割合は小さく，河畔林の硝酸態窒素が流出したに過ぎなかった。土壌水分が増加し，草地−河畔林の連結性が確立されるイベント中には，草地の土壌溶液の河川水への寄与率が増大した。しかし、イベント後期には，硝酸態窒素濃度は低下したことから，脱窒による流出抑制の可能性が示唆された。

窒素流出の時空間変動を SWAT（Soil and Water Assessment Tool）を用いて解析した。これには、90m メッシュの標高数値地図（DEM）、1:50000 土壌図、1:25000 土地利用図、気象データ（5 つの観測地点）、営農関連データ（施肥、放牧、収量）を用いた。標津川で特有の伏流水の影響を加味できるように SWAT を調整したところ，地下水からの窒素負荷は 180 kg d^{-1} と見積もられた。窒素流出は，2004−2006 年のデータを用いて SWAT を校正して得たパラメータ値を用いて，2007 と 2008 年を予測し実測値と比較した。日流量は，計算値と実測値の間にはそれぞれ R^2=0.65, 0.66 の相関があり，よく一致した。月間硝酸態窒素の流出量は，ナッシュ係数（算出値と実測値の適応度合い係数）でそれぞれ 0.89 と 0.72 が認められ，計算値は実測値をよく再現した。さらに、30 年間（1980−2009 年）の窒素流出をこのモデルで見積もったところ，シミュレーションにより、土地利用，地形，土壌タイプ，脱窒，管理の条件に水文過程と生物地球化学的過程が複雑に関わって，硝酸態窒素流出の変動に影響を及ぼすことが示された。

以上のように，本研究は，流域における窒素流出が，明確に時空間スケールで変動しており，農業由来の窒素汚染を効果的に制御するためには，窒素流出のメカニズムやそれに影響する要因の確かな理解に基づく的確な管理手法を構築する必要があることを示したものであり，関連学会からも高く評価されている。よって審査員一同は，Rui Jiang が博士（農学）の学位を受けるのに十分な資格を有するものと認めた。