Assessment of changes in riverine nitrate in the Sesan, Srepok and Sekong tributaries of the Lower Mekong River Basin

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\textbf{Abstract}

Changes in nitrates are of particular concern in tropical regions undergoing rapid development, as these changes may affect local and downstream riverine ecosystems. This study assessed the spatial and temporal differences in nitrate loads within the Sesan, Srepok, and Sekong (3S) Rivers, the largest tributaries of the Mekong River. Simulation results from a flow and nitrate calibrated SWAT model show large differences in year-to-year nitrate loads, a strong seasonality, and clear variability patterns in monthly nitrate loads in the 3S outlet during the wet season. The annual total nitrate loading from the 3S Rivers account for approximately 30% of the total nitrate load of the Mekong River at Pakse. Nitrate loads during the rainy season accounts for 79% of the total annual load into the Mekong River. The Sesan, Sekong, and Srepok basins have average nitrate yields of 400, 330, and 290 kg N/km\textsuperscript{2}, respectively, which is comparable with other forested catchments, but much lower than agriculture dominated catchments in the tropics. Simulations of three future climate scenarios show little variability in annual nitrate loadings under current land use/land cover (LULC), but seasonal difference in nitrate loading during rainy months was observed. Further research is needed to estimate nitrate loads in the 3S basin as influenced by LULC change and dam development, which may potentially result in complex changes to local and downstream riverine ecosystems.

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1. Introduction

Global riverine nutrient inputs into the oceans have tripled during the second half of the 20th century (Jennerjahn et al., 2004). The global biogeochemical cycles of Nitrogen (N) have been significantly altered due to the increasing demand of food and energy consumption caused by increasing population and human activities (Galloway and Cowling, 2002; Seitzinger et al., 2010). The rate at which biologically available nitrogen enters the terrestrial biosphere has more than doubled in the past five decades through activities such as fertilizer production and use, fossil fuel combustion, and cultivation of leguminous crops (Galloway et al., 2004).

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Numerous studies exist on catchment-level changes that influence nutrient cycling and lead to nutrient enrichment, changes in algal and fish ecology, deterioration of water for drinking and recreational purposes, and other consequences (Smith et al., 2003; Jennerjahn et al., 2004). The main sources of high nitrate loadings in river systems are fertilizer application in agriculture, wastewater treatment effluent, and burning processes via deposition of gaseous nitrogen (Zweimüller et al., 2008). Nitrate transport to waterways from diffuse sources is a major cause of eutrophication and episodic acidification for inland aquatic systems and coastal zones (Meader and Goldstein, 2003; Wellington and Driscoll, 2004). High nitrate concentrations in streams and aquifers are also a major concern for drinkable water supplies and for the health of aquatic ecosystems (Gascuel-Odoux et al., 2010).

Climate variability is a strong driver of the hydrological cycle, and therefore it modifies the fate and transport characteristics of nutrients (Bouraoui et al., 2002). Climate is a major driver for biological, chemical and physical processes which determine nitrate cycling (Howden and Burt, 2009; Jones and Smart, 2005; Zhang and Schilling, 2005). Changes in the natural water regime alter the ability of river systems to retain, transform, and transport nutrient loadings originating from upstream and upslope regions (Seitzinger et al., 2002). Climate dictates the seasonality of flows, which controls nutrient export patterns (Pionke et al., 1999). Hydrologically active periods, particularly flood events, are important because the addition of new water sources during such events mobilizes distinctly new and different sources of nutrients from the catchment to the river (Buda and DeWalle, 2009). Oeurung et al. (2010) showed that strong temporal variability of nitrate transport occurred during flood events in a large agricultural catchment in south-west France. A study by Zweimüller et al. (2008) on the effects of climate change on nitrate loads in the Austrian Danube River showed that more nitrate will be transported during winter and less during summer as a direct consequence of temperature change, which is a major driver for biological, chemical and physical processes which determine N cycling and losses (Howden and Burt, 2008). Climate change will also increase the nutrient losses to surface water by accelerating soil processes such as mineralization of organic matter as has been shown in a study of the Yorkshire Ouse catchment in the UK (Bouraoui et al., 2002). In tropical regions of Australia, the effect of agriculture on nitrate yields has also been shown to be significant and nitrates were found to be transported efficiently downstream, with processes such as denitrification in channels being limited (Brodie and Mitchell, 2005). In short, the effect of climate change varies from region to region and the effect of climate on nitrate loads depends on catchment-specific parameters. It is therefore important to understand how climate change affects riverine nitrate loads on a regional and catchment specific basis.

Various models to simulate nitrogen transformation processes, fate, and transport have been developed at the catchment scale to study N dynamics and spatial interactions, including the AGRicultural Non-Point Source Pollution Model (AGNPS), Better Assessment Science Integrating Point and Nonpoint Sources (BASINS), Erosion Prediction Impact Calculator (EPIC), Soil and Water Assessment Tool (SWAT), Hydrological Simulation Program FORTRAN (HSPF), Storm Water Management Model (SWMM), and Water Quality Analysis Simulation Program (WASP) (e.g., Beasley et al., 1980; Duda et al., 2003; Edwards et al., 1994; Arnold et al., 1998; Kinerson et al., Bicknell et al., 2005; Gironás et al., 2010; Di Toro et al., 1983). Among these models, SWAT has been the most widely used to assess hydrology in catchments, as well as to help identify pollution sources (Holvoet et al., 2008), to assess impacts of climate change (Singh and Gosain, 2011), and to assess agricultural management practices (Moriasi et al., 2011). SWAT is considered to be an appropriate tool for assessing nitrate fate from daily to yearly time steps for a wide range of catchment configurations (Santhi et al., 2001; Grizzetti et al., 2003; Jha et al., 2007; Lam et al., 2010; Bothias et al., 2014; Zhai et al., 2014).

Changes in nitrate are of particular concern in tropical regions undergoing rapid development such as the Mekong basin (MRC, 2003; Galloway et al., 2004). The Mekong is the largest river basin in Southeast Asia, covering an area of 795,000 km² where millions of people depend on local fish and rice for their subsistence. Agricultural, ecological, and fish productivity in the lower Mekong, particularly in the Tonle Sap lake in Cambodia and the Mekong Delta in Vietnam, are attributed to the seasonal delivery of water, sediments, and nutrients (Arias et al., 2014a; Kummel et al., 2008; Lamberts, 2006). The Mekong is facing the disruption of its nutrient balance as large increases to nutrient inputs to surface water are expected in the twenty-first century due to increases in agricultural production and infrastructure development (MRC, 2003, 2011; Chea et al., 2016). In southern Vietnam, a significant disturbance to the nitrogen balance of the region has already been observed and it has been attributed to agricultural development in that region (Watanabe et al., 2002). Some large scale studies on nutrients in the Mekong have been conducted (Yoshimura et al., 2009; Liljesthröm et al., 2012; Li and Bush, 2015), but more detailed research on nitrate loadings is needed in key tributaries. Drastic changes in land use, climate, and water infrastructure development occurring in the key Sesam, Srepok, and Sekong (3S) tributaries of the Mekong are of great concern because these rivers’ significant contribution of sediments, nutrients, water flows, and fish diversity to the downstream Tonle Sap Lake and Mekong Delta (Ziv et al., 2012; Arias et al., 2014b). Changes in water flows will be significant due to the future development of over 41 hydropower dams in the 3S basin (Piman et al., 2012) and changes in sediment as also expected to be significant (Kummel et al., 2010; Wild and Loucks, 2014; Kondolf et al., 2014). Information is currently not available as to how hydropower reservoirs operations affect nutrient levels in the Mekong, but it is well known that changes in the nitrogen cycle can occur in large hydropower reservoirs (Kunz et al., 2011). Overall, a quantification of the current nitrate levels along segments of the 3S Rivers is first needed to understand baseline levels, as well as an estimation of potential changes due to climatic change and land use conversion, as these will affect local riverine ecosystems and the provision of aquatic biodiversity, ecosystem services and fisheries to the lower Mekong.

Despite future prospects of climate change, land use conversion, and hydropower development, little is known about historical trends in spatial and temporal variability of nitrate loads from the Mekong tributaries and within the 3S basin.
Table 1
Summary of 3S basin characteristics (CNMC, 2009).

<table>
<thead>
<tr>
<th>Sub basins</th>
<th>Sekong</th>
<th>Sesan</th>
<th>Srepok</th>
</tr>
</thead>
<tbody>
<tr>
<td>Basin characteristics</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Area (km²)</td>
<td>28,815</td>
<td>18,888</td>
<td>30,942</td>
</tr>
<tr>
<td>Elevation range (mamsl)</td>
<td>49–2165</td>
<td>50–2360</td>
<td>49–2358</td>
</tr>
<tr>
<td>Average elevation (mamsl)</td>
<td>543</td>
<td>576</td>
<td>392</td>
</tr>
<tr>
<td>Meteorology based on data from 1980 to 2008</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Average annual temperature range (°C)</td>
<td>26–28</td>
<td>22–27</td>
<td>22–27</td>
</tr>
<tr>
<td>Total average annual precipitation (mm)</td>
<td>2774</td>
<td>2605</td>
<td>2510</td>
</tr>
<tr>
<td>Total wet season precipitation (mm) and percentage (%) of total</td>
<td>2451 (88)</td>
<td>2342 (90)</td>
<td>2142 (85)</td>
</tr>
<tr>
<td>Total dry season precipitation (mm) and percentage (%) of total</td>
<td>323 (12)</td>
<td>263 (10)</td>
<td>368 (15)</td>
</tr>
<tr>
<td>Soil and landuse</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Major soil type and percentage (%) of total basin area</td>
<td>Acrisol (70), Cambisol (14)</td>
<td>Acrisol (79), Ferralsol (14)</td>
<td>Acrisol (60), Ferralsol (20)</td>
</tr>
<tr>
<td>Major land use type and percentage (%) of total basin area</td>
<td>Forest (90), Agriculture (5)</td>
<td>Forest (60), Agriculture (13)</td>
<td>Forest (66), Agriculture (17)</td>
</tr>
</tbody>
</table>

Therefore, the main purpose of this study is to quantify baseline nitrate loads from the 3S tributaries and spatial differences in loadings within the basin using an available set of nitrate concentration samples from each river. Potential changes in spatial and temporal nitrate loads within the 3S tributaries under different climate change scenarios are investigated using the SWAT model. The effect of land use on nitrates is explored spatially using the current spatial distribution of land use/land cover (LULC) in the region.

2. Materials and methods

2.1. Study area

The 3S basin, encompassing the Sesan, Srepok, and Sekong Rivers, consists of a combined catchment area of approximately 78,650 km², covering parts of Cambodia (33% of total basin area), Lao PDR (29%), and Viet Nam (38%; ADB, 2006). The Cambodian part of the basin ranges from the mountains of northeastern Cambodia, which are characterized by rugged terrain with peaks of over 1546 m, to valleys and lowlands. The majority of these areas remain under forest and woodland, with limited agricultural development in the valleys and shifting cultivation on the slopes (CNMC, 2009). The basin also includes part of the Central Highlands of Viet Nam, which are heavily cultivated. The Sekong River flows through Lao PDR before merging with the Sesan and Srepok Rivers in Cambodia. The Sesan and Srepok Rivers flow from Viet Nam to Cambodia where the rivers merge over a distance of about 40 km before the confluence with the main stem of the Mekong River at Stung Treng province of Cambodia (Fig. 1).

The climate in the 3S basin is governed by monsoons. The Southwest Monsoon brings rains in the period from May to October, and the Northeast Monsoon brings dry winds from the Chinese mainland from December to February resulting in a temperature drop and low rainfall (CNMC, 2009). During March and April, hot and dry weather result in particularly high potential transpiration demands. Daily temperature varies between 36 °C, during the hottest months of March/April, to 19 °C during the coldest month of January. Average annual rainfall varies significantly across the subbasins from 1500 mm along the lower reaches of Srepok in the central part of the 3S Basin, to some 2500 mm in the south and more than 3000 mm in the north (ADB, 2006). Rainfall is distinctly seasonal with more than 80% of the annual rain occurring during the rainy season from May to November. The 3S Rivers contribute a combined mean annual discharge of about 2890 m³/s, or about 20% of the Mekong River’s 15,000 m³/s mean annual discharge (MRC, 2005; Adamson et al., 2009). Monsoon-driven discharge (June–November) accounts for about 80% of the annual flow (MRC, 2005). Table 1 provides details on basin characteristics, meteorology, and major soil and land use type for all three basins.

Water quality observations and nitrate load estimation

The Water Quality Monitoring Network Programme under the Mekong River Commission (MRC) was the main source of information on river nitrate data in the 3S. Four years of monthly sampled nitrate concentration (2005–2008) were available at the Lumphat station of the Srepok River, Andoungmeas station of the Sesan River and Siempang of the Sekong River (Fig. 1). Monitoring and analyses were performed by laboratories in Cambodia, under the overall technical guidance of the MRC, which maintains a quality assurance programme. Total nitrite and nitrate were analysed using method 4500–NO2-3/SM from Standard Methods for the Examination of Water and Wastewater (Clesceri et al., 1998). Stations were usually sampled on the 15th of each month, at river depths of 0.3 to 0.5 m in the middle of the cross-sectional profile (thalweg), or at the point of maximum flow if the midpoint was not representative (MRC, 2008a,b). Observed water discharge during the
2005–2008 period was available only for the Lumphat station, but daily flow data for the other sites was obtained from a flow calibrated SWAT model for the basin following the model used by Piman et al. (2012).

Monthly nitrate loads for the monitoring locations were calculated by the product of the measured monthly nitrate concentrations (a sample taken once a month) and the mean monthly water flow at the specific monitoring location. It is important to note that variability of NO₃ differs from suspended sediments in that the concentration levels do not vary significantly on a daily basis within a month in large rivers (Li and Bush, 2015). NO₃ concentrations in the 3S Rivers were found to be very low (majority lower than 1 mg/l) and did not very significantly within a month, as verified by using periods of additional sampled weekly data which showed little variation in concentrations over those periods (Oeurng, 2012).
2.2. Modelling approach

2.2.1. The SWAT model

The Soil and Water Assessment Tool (SWAT 2012) was used to simulate flows and nitrate loadings in the 3S Rivers. SWAT is a physically-based, semi-distributed, agro-hydrological simulation model that operates on a daily time step (as a minimum) at a watershed scale. SWAT is designed to predict the impact of management on water, sediment and agricultural chemical yields in ungauged catchments (Arnold et al., 1998). The model is capable of continuous simulation for dissolved and particulate elements in large complex catchments with varying weather, soils and management conditions over long periods.

SWAT can analyse small or large catchments by discretising into subbasins, which are then further subdivided into hydrological response units (HRUs) with homogeneous land use, soil type and slope. The authors refer to Neitsch et al. (2011) for a detailed description of the model and to Piman et al. (2012) for the application of SWAT to the 3S basin to estimate flow rates. Of particular interest for this study is how SWAT models N. The N cycle of the land phase (i.e. HRU) implemented in SWAT is based on the EPIC model by Williams et al. (1984). SWAT uses a net-mineralisation model for nitrogen, because organic N from the active pool is directly converted to nitrate, whereas gaseous N losses are not modelled explicitly. SWAT monitors five different pools of inorganic N (NH₄⁺ and NO₃⁻) and organic N in the soil. In the HRUs, N is modelled by SWAT in the soil profile and in the shallow aquifer. The N mineralization algorithms in SWAT were adapted from a model for pasture Production limited by rainfall and nitrogen mineralization (Seligman and Van Keulen, 1981). N is divided into soluble and insoluble states and loads are estimated by water volume and average nitrate concentration. SWAT simulates nitrification and ammonia volatilization using a combination of methods (Reddy et al., 1979; Godwin et al., 1984). Nitrate may be transported to streams with surface runoff and lateral flow or percolated to enter the shallow aquifer in recharge from the soil profile. Nitrate in the shallow aquifer may remain in the aquifer, move with recharge to the deep aquifer, move with groundwater flow into the main channel, or be transported out of the shallow aquifer with water moving into the soil zone in response to water deficiencies. Neitsch et al. (2011) also provides a detailed description of the N component simulated in SWAT.

2.2.2. SWAT model input

A digital elevation model, a soils map, and a LULC map were used as key input layers for the SWAT model. A digital elevation model with 250 m resolution for the entire lower Mekong (Fig. 1) was derived from scanned topographical map sheets where contours and spot heights were selectively vectorised by the MRC. The soils map, also created by MRC, was derived from a 2002 soils map using the FAO:UNESCO 1988 soil classification containing 78 soil types (Fig. S1 in Supplementary material). The LULC map was also developed by MRC with 2003 satellite information and included 33 different LULC classes (Fig. S2 in Supplementary material).

Climatological data (temperature, evaporation, humidity, wind speed, and solar radiation) were obtained from six stations within the basin (1980–2008). The precipitation data provided by MRC for the 3S basin are at subbasin levels. MRC uses the MQUAD programme (Hardy, 1971) to interpolate and aggregate the observed precipitation data from stations to the subbasins. MQUAD estimates areal rainfall by calculating a multiquadratic surface from available point rain gauge data, such that the surface passes through all gauge points. For details on MQUAD readers are referred to Shaw and Lynn (1972). The SWAT model used in this study covered an area of 101,414 km², included 140 subbasins, and 2282 HRUs. The model was run on a daily time step for five warm-up years and 29 simulation years (1980–2008). Version 2012 of ArcSWAT (Build 3134, Rev 591) was used in this study.

2.3. Model calibration and validation

The SWAT model was calibrated and validated independently for water discharge and then for nitrate loads. Daily water discharges were calibrated (1985–2000) and validated (2001–2005) at eight different river flow monitoring stations (MRC, 2011). The parameters for flow simulations were fitted through an auto calibration procedure using SWAT-CUP for the 8 river flow stations using meteorology data from the Mekong River Commission. Nitrate calibration based on monthly loads from 2005 to 2008 was also carried out using a sequential uncertainty fitting algorithm (SUFI-2) with SWAT-CUP (Abbaspour, 2013). The initial parameter ranges for optimization were based on the likely maximum range recommended for each parameter by the SWAT and SWAT-CUP developers for the conditions in the basin (Table 2). The Nash-Sutcliffe model efficiency factor (NS) was used as the objective function in the nitrate calibration because it has been shown to be a robust objective function that reflects the overall fit of hydrographs (Servat and Dezetter, 1991) and it is widely used for water quality calibrations (Gupta et al., 2009). It ranges from minus infinity to one, where one represents a perfect model match. In order to test the sensitivity of nitrate load estimates to the different calibration parameters, a global sensitivity analysis was carried out with SWAT-CUP on the final set of calibrated parameters, which included 500 different parameter combinations. Table 2 shows the fitted values of parameters used for calibration of nitrate load.

The model’s performance was evaluated by comparing the simulated with the observed constituents using NS and Coefficient of determination (R²). A calibrated model could be judged satisfactory if NS and R² values are >0.6 for mean behaviour (Benaman et al., 2005). This threshold, however, is rather subjective and should be used with caution and with consideration of the model’s objective variable.
Table 2
Fitted parameter values for the 3S basin and initial parameter ranges calculated using SUFI-2 during calibration of Nitrate.

<table>
<thead>
<tr>
<th>Parameter name</th>
<th>Definition</th>
<th>Minimum value</th>
<th>Maximum value</th>
<th>Fitted value</th>
</tr>
</thead>
<tbody>
<tr>
<td>r_ERORGN.hru</td>
<td>Organic N enrichment for sediment</td>
<td>2.217</td>
<td>6.653</td>
<td>4.453</td>
</tr>
<tr>
<td>v_RSDCO_bsn</td>
<td>Residue decomposition coefficient</td>
<td>0.037</td>
<td>0.079</td>
<td>0.058</td>
</tr>
<tr>
<td>v_SOLNO3_chm</td>
<td>Initial NO₃ concentration in the soil layer (mg N/kg soil, dry weight)</td>
<td>39.940</td>
<td>119.859</td>
<td>79.90</td>
</tr>
<tr>
<td>v_CMN_bsn</td>
<td>Rate factor for humus mineralization of active organic nutrients</td>
<td>0.000039</td>
<td>0.002</td>
<td>0.001</td>
</tr>
<tr>
<td>r_SHALLST_N_gw</td>
<td>Nitrate concentration in shallow aquifer</td>
<td>298.403</td>
<td>895.597</td>
<td>597.000</td>
</tr>
<tr>
<td>r_A11_wwq</td>
<td>Fraction of algal biomass that is nitrogen</td>
<td>0.078</td>
<td>0.929</td>
<td>0.853</td>
</tr>
<tr>
<td>v_BC2_BSN_bsn</td>
<td>Rate constant for biological oxidation for nitrogen in reach</td>
<td>−0.217</td>
<td>1.261</td>
<td>0.522</td>
</tr>
<tr>
<td>v_CH_ONCO_re</td>
<td>Organic nitrogen concentration in the channel</td>
<td>−23.459</td>
<td>58.859</td>
<td>17.700</td>
</tr>
<tr>
<td>v_SOLORGN_chm</td>
<td>Initial organic N concentration in the soil layer (mg N/kg soil, dry weight)</td>
<td>−35.459</td>
<td>54.859</td>
<td>9.700</td>
</tr>
<tr>
<td>v_NPERCO_bsn</td>
<td>Nitrate percolation coefficient</td>
<td>−0.144</td>
<td>0.618</td>
<td>0.237</td>
</tr>
<tr>
<td>v_LAT_ORGN_gw</td>
<td>Organic N in baseflow (mg/l)</td>
<td>−66.738</td>
<td>111.138</td>
<td>22.200</td>
</tr>
</tbody>
</table>

*Note:* a The extension (e.g., hru) refers to the SWAT input file where the parameter occurs; b The qualifier (v) refers to the substitution of a parameter by a value from the given range; and c The qualifier (r) refers to relative change in the parameter where the value from the SWAT database is multiplied by 1 plus a factor in the given range.

SUFI-2 was also used for the uncertainty analysis of the SWAT model. In SUFI-2, the degree to which uncertainties are accounted for is quantified by the p-factor and the r-factor. The p-factor is the percentage of measured data bracketed by 95% prediction uncertainty (95 PPU). The r-factor is the average thickness of the 95 PPU band divided by the standard deviation of the measured data. The goodness of fit and the degree to which the calibrated model accounts for the uncertainties are assessed by the closeness of the p-factor to 100% (i.e., all observations falling inside the prediction uncertainty band) while having the narrowest band (r-factor → 0).

### 2.4. Climate scenarios and downscaling

A dataset of downscaled global climate change scenario (IPCC 5th Assessment Report) were obtained from the MRC Climate Change and Adaptation Initiative (CCAI). This dataset includes SWAT model monthly ‘change factors’ (i.e., the magnitude of relative changes with respect to the baseline/historical conditions of each global circulation model [GCM]) for precipitation, temperature, solar radiation and relative humidity. MRC CCAI uses SimCLIM software to downscale the climate. SimCLIM uses pattern scaling plus bilinear interpolation algorithm to downscale the GCM outputs. MRC CCAI uses change factors to quantify the projected alterations to the climate because the change factor approach represents the simplest and most practical way to produce scenarios based on multiple GCMs, emission scenarios, sensitivities, time horizons and locations (MRC, 2015).

Large uncertainty can result from the selection of different GCMs (MRC, 2015), and therefore, the procedure needs to be carefully carried out. Three GCMs (GISS-E2-R-CC, IPSL-CM5-MR and GFDL-CM3) and Representative Concentration Pathway (RCP) 6.0 were considered for general impacts of climate change in Lower Mekong Basin based on the recommendations from the study by MRC (2015). GISS-E2-R-CC projects a ‘drier’ future on average across all locations and seasons (i.e. GISS-E2-R-CC projects less rain in most of the MRB, but there are some local departures from this pattern (MRC, 2015)). IPSL-CM5A-MR is the ‘medium’ scenario, since it projects wetter wet seasons and drier dry season (i.e. increased seasonal variability). GFDL-CM3 represents the ‘upper’ bound of projected future impacts (i.e. wetter overall).

Three time horizons (near-term future 2021–2040, medium-term future 2051–2070, long-term future 2081–2100) were considered in this study.

## 3. Results

### 3.1. Water discharge and nitrate simulation

Calibration and validation of daily water discharge show an overall increase in model fitting from upstream to downstream monitoring stations (Table 3). NS coefficient of efficiency varied from 0.41–0.42 in the upper Sesan at Kontum to 0.96–0.97 in the Mekong at both Stung Treng and Kratie. Differences in the total computed water volume versus the total observed volume were generally low, ranging from 1 to 21% at Kontum to 0–1% at Stung Treng.

Calibration of nitrate loads for monthly estimates was carried out at three of the monitoring stations for which multiple years of continued nitrate sampling had taken place. The calibration revealed that the model adequately estimated seasonal
Fig. 2. Nitrate load calibration results for monthly estimates (2005–2008) at (A) Siempang on the Sesan River, (B) Andaungmeas on the Sesan River, (C) Lumphat on the Srepok. The solid shading (95PPU) represents 95% of the prediction uncertainty for 11 parameter combinations resulting in NS coefficients for all monitoring stations presented in Table 4.
Table 3  
Calibration and validation of water discharge.

<table>
<thead>
<tr>
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<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td>NS coefficient of efficiency</td>
<td>NS coefficient of efficiency</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>Vol ratio (computed/observed)</td>
<td>Volume ratio (computed/observed)</td>
</tr>
<tr>
<td>Kratie</td>
<td>Mekong</td>
<td>13040</td>
<td>0.97</td>
<td>0.96</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>1.00</td>
<td>0.99</td>
</tr>
<tr>
<td>Stung Treng</td>
<td>Mekong</td>
<td>12548</td>
<td>0.97</td>
<td>0.96</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>1.01</td>
<td>1.00</td>
</tr>
<tr>
<td>Lumphat</td>
<td>Srepok</td>
<td>740</td>
<td>0.60</td>
<td>0.59</td>
</tr>
<tr>
<td>Attapeu</td>
<td>Sekong</td>
<td>426</td>
<td>0.54</td>
<td>0.64</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>1.01</td>
<td>0.95</td>
</tr>
<tr>
<td>Bandon</td>
<td>Srepok</td>
<td>278</td>
<td>0.64</td>
<td>0.53</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>1.03</td>
<td>1.12</td>
</tr>
<tr>
<td>Cau 14</td>
<td>Srepok</td>
<td>250</td>
<td>0.63</td>
<td>–</td>
</tr>
<tr>
<td>Trung Nghai</td>
<td>Sesan</td>
<td>132</td>
<td>0.47</td>
<td>–</td>
</tr>
<tr>
<td>Kontum</td>
<td>Sesan</td>
<td>96</td>
<td>0.41</td>
<td>0.42</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>1.01</td>
<td>1.21</td>
</tr>
</tbody>
</table>

Table 4  
Nitrate load calibration results.

<table>
<thead>
<tr>
<th>Station name: River: Objective function</th>
<th>Monitoring station 1</th>
<th>Monitoring station 2</th>
<th>Monitoring station 3</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Siempang</td>
<td>Andaungmeas</td>
<td>Lumphat</td>
</tr>
<tr>
<td></td>
<td>Sekong Calibration fit</td>
<td>Sesan Calibration fit</td>
<td>Srepok Calibration fit</td>
</tr>
<tr>
<td>p-factor</td>
<td>0.25</td>
<td>0.52</td>
<td>0.29</td>
</tr>
<tr>
<td>r-factor</td>
<td>0.50</td>
<td>0.52</td>
<td>0.38</td>
</tr>
<tr>
<td>R²</td>
<td>0.70</td>
<td>0.71</td>
<td>0.57</td>
</tr>
<tr>
<td>NS</td>
<td>0.58</td>
<td>0.67</td>
<td>0.49</td>
</tr>
</tbody>
</table>

Fig. 3. Variability in monthly river nitrate yields at the 3S outlet for 21 years of simulation.

variations in nitrate loads (Fig. 2). R² for the calibration period was 0.57 to 0.71 for all three stations, while NS varied from 0.49 to 0.67 (Table 4).

3.2. Seasonal and temporal variability of nitrate load transport

As expected, monthly nitrate loads in the 3S rivers outlet showed a strong seasonality and clear variability patterns (Fig. 3). Monthly loads during the rainy season months of July, August, September and October were 7.9 (SD ± 2.5), 10.8 (SD ± 2.4), 10.9 (SD ± 1.8) and 8.9 (SD ± 1.3) × 10³ t, respectively. The total export of nitrate loads during the rainy season accounts for 78% of the total annual load from the 3S into the Mekong River. In contrast, both magnitude and variability are low during the dry season months of January, February, and March.

A 21-year baseline series (1985–2005) at the Sekong, Sesan, Srepok and the 3S outlet showed a large difference in year-to-year nitrate loads, assuming no change in LULC through time (Fig. 4). The nitrate load ranged from 16 to 32.1 × 10³ t/year (mean: 27 × 10³ t) for the Sekong River, 9 to 15.8 × 10³ t/year (mean: 13 × 10³ t) for the Sesan River, and 13.5 to 25.3 × 10³ t/year (mean: 18.4 × 10³ t) for the Srepok River. The annual total nitrate load from the 3S rivers to the Mekong River varied between 44.7 and 74.2 × 10³ t/year (mean: 57.6 × 10³ t). Even though the spatial variability of riverine nitrate loads at different reaches in the 3S river basins follow the magnitudes of flow, the spatial variability of nitrate concentrations indicates a more complex relationship as a function of HRU’s characteristics (Fig. 5A & B).

The annual nitrate load from the model was significantly correlated with annual water discharge for the 3S outlet, with an R² value of 0.93 (Fig. 6). Based on this strong relationship, annual water discharge could be used to estimate annual nitrate
load for long-term periods within the 3S catchment, but only under conditions where current land use does not change drastically. However, the flow vs. nitrate load correlation is not consistent within the 3S subbasins; the Sesan and Srepok have very high correlations whereas the Sekong has a lower correlation (Fig. 6). The Sekong shows lower correlation values than the Sesan or Srepok basins due to less nitrate input (large forest areas) with large water flows.

3.3. Spatial patterns of subbasin nitrate yield

Large spatial variability in nitrate yields occurred at the subbasin level in the study area (Fig. 5A). The average nitrate yield in the whole 3S was an estimated 330 kg/km²; however, some agricultural subbasins have nitrate yields higher than 1200 kg/km² and some forested subbasins have yields as low as 135 kg/km². A plot of nitrate yield versus percentage of agriculture in both lowland and upland subbasins shows a general trend of higher nitrate yields as percent agriculture in the subbasin increases (Fig. 7). The Sesan Basin has the highest nitrate yield with an average of 400 kg/km², the Sekong yields 330 kg/km², and the Srepok has the lowest average yield of 290 kg/km². As expected, the subbasins with higher agriculture land use tend to have higher nitrate yields than those with forest land as can be witnessed by the higher yields from Vietnam where most of the land is under agricultural production, but other factors also come into play. For instance, a subbasin with agricultural land of 44% of the total subbasin in the Sekong has nitrate yields of up to 848 kg/km², whereas in the Sesan basin, a subbasin nitrate yield of 1619 kg/km² can be found where crops accounted for only 43% of the total subbasin land area. A relationship of land use type to downstream nitrate concentrations in river reaches can also be inferred (Fig. 5B), where upstream agricultural land use results in higher riverine nitrate concentrations. Other factors such as spatial variability in terrain (elevation/slope) and rainfall also have an effect on nitrate yields (numbered item in Figs. 7 and 5A), although the effect of these is less obvious given the relatively low concentrations observed.

3.4. Effects of climate change on temporal and seasonal variability of nitrate load

The results from our simulations revealed only relatively small differences in interannual variability of nitrate loads between baseline and climate change simulations (Fig. 8A). The simulation of the GISS climate scenario predicted a nitrate load increase by 9% compared to the baseline load for the 2021–2040 period, but a decrease 1.8 to 2.4% for 2051–2070 and 2081–2100, respectively (Table 5). Nitrate loads under the GFDL-CM3 and IPSL-CMS-MR climate scenarios are 1.1 to 5.4% higher than the baseline nitrate for the three time horizons. Annual spatial patterns of nitrate yields under the various climate scenarios were not significantly different from baseline values and resulting maps were similar to the one presented in Fig. 5A. Boxplots in Fig. 8B show a relatively similar distribution of loads among scenarios and a discrepancy in the direction of changes for the future scenarios. While GISS shows a small reduction in mean loads, the other two scenarios show a tendency to increase. In particular, the GFDL-CM shows the greatest increase in mean loads and also a small tendency
Table 5
Percentage of change in mean annual flow and nitrate load transport for different climate scenarios compared with baseline.

<table>
<thead>
<tr>
<th>Scenario</th>
<th>2021-2040 % Change</th>
<th>2051-2070</th>
<th>2081-2100</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Flow</td>
<td>NO₃</td>
<td>Flow</td>
</tr>
<tr>
<td>GISS</td>
<td>11.7</td>
<td>9.0</td>
<td>−4.2</td>
</tr>
<tr>
<td>GFDL-CM3</td>
<td>0.8</td>
<td>1.6</td>
<td>2.7</td>
</tr>
<tr>
<td>IPSL-CM5A-MR</td>
<td>0.6</td>
<td>1.1</td>
<td>0.8</td>
</tr>
</tbody>
</table>

Seasonal variability of nitrate loads, however, was observed between the baseline and climate change scenarios for the wet months of the year (Fig. 9 and Table 6). Changes in nitrate loads during dry months were not significant. Under the GISS climate scenario, nitrate load increased by 49.4% in May for the 2021–2040 period, but decreased by 23.5% for 2051–2070 and by 31.4% for 2081–2100. Under the GFDL-CM3 climate scenario, nitrate load increased by 4.8% for 2021–2040, 8% for 2051–2070 and 10.7% for 2081–2100, respectively. Nitrate loads decreased under the IPSL-CM5A-MR scenario in the early wet season for all three time horizons.

to increase variability. Despite the visible difference among scenarios, none of the future scenarios are significantly different from the baseline.

Seasonal variability of nitrate loads, however, was observed between the baseline and climate change scenarios for the wet months of the year (Fig. 9 and Table 6). Changes in nitrate loads during dry months were not significant. Under the GISS climate scenario, nitrate load increased by 49.4% in May for the 2021–2040 period, but decreased by 23.5% for 2051–2070 and by 31.4% for 2081–2100. Under the GFDL-CM3 climate scenario, nitrate load increased by 4.8% for 2021–2040, 8% for 2051–2070 and 10.7% for 2081–2100, respectively. Nitrate loads decreased under the IPSL-CM5A-MR scenario in the early wet season for all three time horizons.

Fig. 5. Spatial variability of mean nitrate loads and yields (A) and concentrations (B) within different reaches of the 3S Rivers. Numbered subbasins in (A) are ones with nitrate yields greater than 800 kg/km² (see also Fig. 7).
Table 6
Percentage change in monthly nitrate load transport for different climate scenarios compared with baseline for the periods 2021–2040 and 2081–2100.

<table>
<thead>
<tr>
<th>Month</th>
<th>2021–2040</th>
<th></th>
<th>2081–2100</th>
<th></th>
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</thead>
<tbody>
<tr>
<td></td>
<td>GISS</td>
<td>GFDL-CM3</td>
<td>IPSL-CMSA-MR</td>
<td>GISS</td>
</tr>
<tr>
<td>Jan</td>
<td>14.2</td>
<td>3.0</td>
<td>4.6</td>
<td>2.7</td>
</tr>
<tr>
<td>Feb</td>
<td>16.2</td>
<td>1.5</td>
<td>3.0</td>
<td>2.9</td>
</tr>
<tr>
<td>Mar</td>
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<td>3.2</td>
<td>−0.7</td>
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<tr>
<td>Apr</td>
<td>52.0</td>
<td>5.1</td>
<td>−15.6</td>
<td>−15.9</td>
</tr>
<tr>
<td>May</td>
<td>49.4</td>
<td>−3.4</td>
<td>−22.7</td>
<td>−31.4</td>
</tr>
<tr>
<td>Jun</td>
<td>16.8</td>
<td>1.1</td>
<td>−7.7</td>
<td>−23.4</td>
</tr>
<tr>
<td>Jul</td>
<td>6.6</td>
<td>−1.4</td>
<td>−0.5</td>
<td>−6.7</td>
</tr>
<tr>
<td>Aug</td>
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<td>0.1</td>
<td>1.4</td>
<td>4.3</td>
</tr>
<tr>
<td>Sep</td>
<td>6.4</td>
<td>4.8</td>
<td>4.5</td>
<td>7.0</td>
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<tr>
<td>Oct</td>
<td>1.7</td>
<td>1.2</td>
<td>4.1</td>
<td>−3.1</td>
</tr>
<tr>
<td>Nov</td>
<td>5.2</td>
<td>2.1</td>
<td>4.9</td>
<td>−3.3</td>
</tr>
<tr>
<td>Dec</td>
<td>12.6</td>
<td>4.5</td>
<td>5.6</td>
<td>0.7</td>
</tr>
</tbody>
</table>
4. Discussion

The temporal variability of nitrate loads in the 3S Rivers is correlated with river flow (Fig. 4). For instance, the lowest annual nitrate load at the 3S outlet over a 21 year period was $44.7 \times 10^3$ t with a mean annual water discharge of 2222 m$^3$/s observed in 1992 while the highest load was of $74.2 \times 10^3$ t with a mean annual water discharge of 3991 m$^3$/s, observed in 1999. Nitrate loads in the Sekong River were 1.8 times that of the Sesan River and 1.3 that of the Srepok River, which is mainly attributed to the higher annual flow of 1167 m$^3$/s of the Sekong River compared to 743 m$^3$/s and 1205 m$^3$/s for the Sesan and Srepok rivers, respectively. The long-term average nitrate load from the 3S to the Mekong River was estimated to be $57.6 \times 10^3$ t year$^{-1}$. This value is about 30% of the total annual nitrate load of the Mekong River at Pakse ($3.3 \times 10^9$ mol year$^{-1}$ or $236 \times 10^3$ t year$^{-1}$) which has been recently reported (Li and Bush, 2015).

It is likely that major future LULC changes in the 3S basins driven by deforestation of native rain forest, expansion of agricultural and urban areas, and expansion of commercial plantations such as rubber trees will cause the greatest change in future nitrate loads (Takamatsumi et al., 2014). Future trends in nitrate loads are not likely to be driven only by environmental change, but mainly by nitrogen sources such as fertilizer additions in cultivated areas (Martinková et al., 2011). In the last few decades, fertilizer application in the Cambodian region of the 3S basins was not widespread among farmers, but this is likely to change in order to achieve higher agricultural yields. High levels of fertilizer applications are already occurring in Vietnam, and Lao PDR will probably follow.

The annual mean nitrate yield of the 3S basins is about $330$ kg km$^{-2}$ yr$^{-1}$, which is comparable with other tropical catchments under mostly natural forest conditions (ie.351 ± 62 kg km$^{-2}$ yr$^{-1}$ for a catchment in Taiwan reported by Huang et al. (2012)). However, this value is much lower than agriculture dominated catchments around the world (Table 7). For
moderately cultivated catchments, the annual reported yields ranged from 628 to 6526 kg km\(^{-2}\) yr\(^{-1}\) with an average of 2327 kg km\(^{-2}\) yr\(^{-1}\) for the reported catchments (Huang et al., 2012). The nitrate yields of 848 kg km\(^{-2}\) to 1619 kg km\(^{-2}\) in some 3S subbasins where agriculture is significant are rather low, but within the value ranges of the moderately cultivated catchments. This means that the sources of nitrates are still low from agricultural activities in the 3S basins. Ranges from 28 988 to 48 914 kg km\(^{-2}\) yr\(^{-1}\) have been reported in eastern central Illinois (USA) for intensive agriculture where almost the whole watershed is under row crop production, primarily corn rotated with soybean and fertilizer applications are significant.

Climate variability affects the hydrological cycle directly, thus modifying the transformation and transport characteristics of nutrients. Changes in nitrate loads in the 3S basins as a result of climate change scenarios are driven mainly by changes in precipitation, temperature (and consequent changes of the form of precipitation) and rainfall intensity. No significant differences were found between baseline average annual nitrate loads and climate change scenario simulations. Seasonal differences, however, were clearly distinguished. The seasonal nitrate loading under the IPSL-CM5A-MR climate scenario for the three time horizons (2030s, 2060s and 2090s) at the beginning of the rainy season was lower than the baseline, but was higher from August when water discharge peaks. There was also an observable increase in loadings under the GISS climate scenario for the near term 2030 period, but a decrease in long-term future (2060s and 2090s). This can be explained by lower rainfall occurring during the early rainy season under climate change in the long term. Other climate change studies in the

Fig. 7. Relationship between average annual nitrate yields and% agriculture in upland (hilly/mountainous) subbasins (A) and lowland (plains) subbasins (B). Numbered points (>800 kg/km\(^2\)) shown in Fig. 5A.
Fig. 8. Temporal variability of nitrate load transport (A) and box plot (B) for the baseline and climate change scenarios at the 3S outlet for the 2081 to 2100 period. The box plot centre line represents the mean (50th percentile), the lower edge the 25th percentile, and the upper edge the 75th percentile level. Minimum and maximum values are represented by error bars.

Fig. 9. Seasonal variability of nitrate load (with 10% error) for the baseline and climate change scenarios at the 3S outlet for the 2081–2100 period.
Lower Mekong Basin have also reported that there will be changes in seasonal distribution of rainfall, with drier and longer dry seasons, and shorter, more intense wet seasons (MRC, 2010; Piman et al., 2015; Kingston et al., 2011; Lauri et al., 2012). Volume and intensity of wet-season rainfall would increase, leading to increased probability of flooding extent, a marginal decrease in dry-season rainfall (Clausen, 2009), and increased frequency and intensity of extreme events, such as floods and droughts.

Dams are known to affect nutrient loads in rivers (Bosch, 2008). Water impoundments by dam construction increase both depth and residence times compared to pre-dam conditions. Phosphorus, which is mainly attached to soil particles in flow, is heavily influenced by trapping of sediment in reservoirs. Nitrates, on the other hand, are transported in their dissolved form in the river water, but can be influenced by dam reservoirs through other biogeochemical means. Increased water retention times from reservoir operations, leading to a reduction in Redox potential and decreased temperatures, will affect nitrate loadings and conversions to other forms of Nitrogen. A recent study on the effects of dams on nutrients in the Huron and Raisin Rivers, for instance, demonstrated that dams played a vital role in effectively removing excess nutrients (Bosch, 2008). This same study showed that dams had the greatest impact when they were placed near the river mouths or in higher nutrient source areas, and when dams were (experimentally) removed from the rivers (through modelling with SWAT), the amount of nitrogen and phosphorus in the rivers doubled. Results from Bosch (2008) imply that the impact on nutrient loadings of the future development of 41 dams in the 3S basin (Piman et al., 2015) could be significant, with unclear resulting ecological positive or negative effects. The interaction between future land use change and hydropower dam development in the 3S in terms of nutrient loadings in riverine systems will be complex and needs further study.

It is also important to note that the assessment of changes in riverine nitrate loading in the 3S basin achieved in this study was based on a limited set of data which spanned only 4 years of monthly water sampling at 3 stations. Given the size of the 3S basins, Continuation and refinement of the basin monitoring network is necessary to improve assessments and future modelling work.

5. Conclusions

This study provides a benchmark for quantifying nitrate loads in the 3S Rivers and an insight into the effects of climatic change on future loads. Nitrate loads were found to be the highest for the Sekong River followed by the Sesan River and then the Srepok River, which is analogous to their respective flow rates. Average nitrate yields, on the other hand, were the highest at the Sesan followed by the Sekong and then the Srepok basin, which closely relates to the agricultural development in each basin. The current nitrate yield from the 3S basin is comparable with forested catchments around the world and much lower than other agriculture dominated catchments.

Simulation with inputs from climate change scenarios had little influence on nitrate loadings in an annual basis for all three climate scenarios simulated, but small seasonal variations were observed between scenarios. Overall, climate change under the current LULC conditions did not have a significant influence on mean nitrate loads at the basin scale. The future nitrate loads in the 3S catchments will be influenced by land use change, which is likely to increase nitrate levels more than climate change as more land is used for agriculture, particularly due to increases in fertilizer use. The future development of 41 dams in the 3S basin will probably cause a change in nutrient dynamics and loads through water impoundments. The combined effect of agricultural land development and hydropower development needs further study, as complex interactions could cause significant changes in riverine nutrient levels, which can affect downstream ecosystems within the basin and beyond.

Acknowledgements

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Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at http://dx.doi.org/10.1016/j.ejrh.2016.07.004.

References


