First Assessment of Inorganic Nitrogen Deposition Budget Following the Impoundment of a Subtropical Hydroelectric Reservoir (Nam Theun 2, Lao PDR)

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Journal of Geophysical Research: Atmospheres

RESEARCH ARTICLE
10.1029/2018JD029027

Key Points:
- Dry and wet inorganic nitrogen deposition fluxes have been estimated for the Nam Theun 2 Reservoir and the vegetated area present before impoundment
- Wet deposition was the major source of the atmospheric nitrogen input to the Nam Theun 2 Reservoir, contributing to 80% of the total deposition
- The total inorganic N deposition over the studied area has been reduced by 26% after impoundment

Supporting Information:
- Supporting Information S1

Correspondence to:
M. Adon and C. Galy-Lacaux,
adonatma@yahoo.fr;
lacc@aero.obs-mip.fr

Citation:
Adon, M., Galy-Lacaux, C., Serça, D., Guedant, P., Vongkhamsao, A., Rode, W., et al. (2018). First assessment of inorganic nitrogen deposition budget following the impoundment of a subtropical hydroelectric reservoir (Nam Theun 2, Lao PDR). *Journal of Geophysical Research: Atmospheres*, *123*, 12,413–12,428. https://doi.org/10.1029/2018JD029027

Received 18 MAY 2018
Accepted 1 OCT 2018
Accepted article online 15 OCT 2018
Published online 8 NOV 2018

First Assessment of Inorganic Nitrogen Deposition Budget Following the Impoundment of a Subtropical Hydroelectric Reservoir (Nam Theun 2, Lao PDR)

Marcellin Adon[1,2], Corinne Galy-Lacaux[2], Dominique Serça[2], Pierre Guedant[3], Axay Vongkhamsao[3], Wanidaporn Rode[3], Yves Meyerfeld[2], and Frédéric Guérin[4,5]

[1]Laboratoire de Physique de l'Atmosphère et de Mécanique des Fluides, Université Félix Houphouët-Boigny, Abidjan, Côte d'Ivoire, [2]Laboratoire d'Aérologie, Université de Toulouse, CNRS, UPS, Toulouse, France, [3]Environment Department, Environment and Social Unit, Nam Theun 2 Power Company Limited, Vientiane, Lao PDR, [4]Université de Toulouse, UPS, GET, Toulouse, France, [5]IRD, UR 234, GET, Toulouse, France

Abstract With around 490 km^2 at full level of operation, the Nam Theun 2 Reservoir (NT2R) is one of the largest hydroreservoir in Southeast Asia. This study presents a first estimation of the atmospheric inorganic nitrogen deposition into the NT2R based on a 2-year monitoring (June 2010 to July 2012) including gas concentrations and precipitation. Dry deposition fluxes are estimated by the inferential method using, on the one hand, surface measurements of gas concentrations (NO_2, HNO_3, NH_3) from passive samplers and, on the other hand, modeled exchange rates. Wet deposition fluxes are calculated from NH_4^+ and NO_3^- concentrations determined in samples of rain from an automatic precipitation collector. The average nitrogen deposition flux is estimated at 1.26 ± 0.14 kgN·ha^{-1}·year^{-1} from dry processes and 5.01 ± 0.92 kgN·ha^{-1}·year^{-1} from wet ones, that is, an average annual total nitrogen flux of 6.27 ± 1.06 kgN·ha^{-1}·year^{-1} deposited at the NT2R with 80% from wet deposition. Before impoundment, the mean N flux has been estimated at 3.42 ± 1.88 kgN·ha^{-1}·year^{-1} for dry deposition and 5.01 ± 2.12 kgN·ha^{-1}·year^{-1} for wet deposition, or a total N deposition flux of 8.43 ± 4.01 kgN·ha^{-1}·year^{-1} over the studied area dominated by forests with little agriculture soil and water surfaces. Thus, the total N deposition over the studied area has been reduced of 26% (or 63% for dry deposition) following the reservoir impoundment based on our working hypothesis.

1. Introduction

Studies of N input to various water surfaces (coastal, lake, river, and ocean) indicate that atmospheric nitrogen deposition represents a significant amount of the total nitrogen input (Canham et al., 2012; Gao et al., 2007; Jung et al., 2009; Qi et al., 2013; Whitall et al., 2003). Recent N deposition-monitoring studies also showed very high N deposition rates at several observation sites including agricultural catchments in different regions of East Asia (Pan et al., 2012; Shen et al., 2013; Sugimoto & Tsuboi, 2017). Although effects are usually the greatest near the sources of pollution (Fenn et al., 2003), atmospheric deposition of N is of particular concern since anthropogenic N deposition can be seen in areas located far from pollution sources (Elser et al., 2009). Therefore, the increase of N deposition is a local, regional, and continental issue. Atmospheric nitrogen deposition can affect the terrestrial ecosystems but also marine systems and inland water, eventually leading to eutrophication and acidification (Goulding et al., 1998; Jones et al., 2014; Liu et al., 2010, St-Laurent et al., 2017). Although extensive work has been done on the effects of N deposition in polluted regions, little is known about N deposition and its impact following the modification of the land use resulting in the conversion of a forested area to a water body. This study is a first attempt to quantify the net perturbation of N deposition following the transfer of a terrestrial landscape into an aquatic system.

Atmospheric N can affect water surfaces directly through wet deposition (precipitation) and dry deposition of both aerosol particles and gases, as in all surfaces. Wet and dry deposition can also indirectly contribute to N budget in water bodies. This is the case when nitrogen collected in the watershed is being delivered to the water body by groundwater and surface runoff. A large fraction of the atmospheric nitrogen input is in the form of inorganic nitrogen, mainly ammonium (NH_4^+) and nitrate (NO_3^-; Ayars & Gao, 2007). Atmospheric

NH_4^+ is derived from heterogeneous reactions involving ammonia (NH_3), while NO_3^- is a gaseous oxidation product of NO_x (NO_2, NO) formed in the atmosphere (Seinfeld & Pandis, 2006). Major sources of NH_3 are linked with livestock farming and emission from natural or fertilized soils (Bouwman et al., 2002) though fossil fuel-based NH_3 emissions can be important in urban areas, especially during periods of haze pollution (Pan et al., 2016). Emission sources for NO_x include combustion of fossil fuels, biomass burning, biogenic soil emissions, and lightening (Delmas et al., 1997, 2005; Galanter et al., 2000; Yienger & Levy, 1995).

Dissolved organic nitrogen (DON) can also contribute to the total N deposition budget with a sizable proportion that can reach up to 10–40% of the wet deposition flux (Cornell et al., 2003; Hill et al., 2005; Nakamura et al., 2006; Whitall et al., 2003; Zhan et al., 2017). Dry deposition of particulate inorganic nitrogen (pNH_4^+, pNO_3^-) can also be nonnegligible in the total N deposition budget over water surfaces (Ayars & Gao, 2007; Gao, 2002). For example, particulate N species contributed approximately 10% to the total N deposition flux over North China (Pan et al., 2012).

This paper presents the estimation of the atmospheric inorganic nitrogen deposition before and after the Nam Theun 2 hydroelectric Reservoir was flooded. The gaseous N concentrations (NH_3, HNO_3, and NO_2) and inorganic N concentrations in rain samples (NH_4^+ and NO_3^-) were measured in the forest surrounding the reservoir for a 2-year period (June/July 2010 to July 2012). Dry deposition fluxes were estimated using the inferential method combining gaseous concentration measurements and modeling of deposition velocities according to the resistance analogy (Wesely, 1989; Zhang et al., 2003). Wet deposition fluxes were estimated from NH_4^+ and NO_3^- concentrations in single-event rain samples. The nitrogen deposition budget has also been calculated before impoundment over the studied area characterized by contrasting land use type (different types of forest, rice paddies, and river surface/wetlands). The perturbation of the nitrogen cycle resulting from the flooding of the reservoir will ultimately be discussed by comparing nitrogen deposition before and after the impoundment.

2. Materials and Methods

2.1. Site Description

The Nam Theun 2 Dam (17°59′49″N, 104°57′08″E) was built on the Nam Theun River, in the subtropical region of the Lao PDR (People's Democratic Republic) and is operated by the Nam Theun 2 Power Company (NTPC). The Nam Theun 2 Reservoir (NT2R) was flooded in 2008, reached it maximum water level in October 2009, and was commissioned in April 2010. This is a transbasin hydroelectric reservoir that diverts water from the Nam Theun River (a Mekong tributary) to the Xe Bangfai River (another Mekong tributary). The reservoir was described in detail in Descloux et al. (2016), and the carbon cycle and especially the greenhouse gas emissions were monitored since impoundment (Chanudet, Guédant, et al., 2016; Chanudet, Smits, et al., 2016; Deshmukh et al., 2014, 2016; Guérin et al., 2016; Serça et al., 2016). The reservoir surface fluctuated from 140 km^2 (525.5 m above sea level) to around 490 km^2 at the full water level (538 m above sea level) between 2009 and 2013. The land use of the flooded area was mainly dense, medium, light, and degraded forests (83%), whereas agricultural lands accounted for 11%, with the rest made up mainly by water surfaces (6% for rivers and swamps) and other types of surface (~1% for roads, villages …; Figure 1; Descloux et al., 2011).

The study site is under a subtropical monsoon climate, with a pronounced warm wet season from May to October, a cool dry season from November to February, and a warm dry season in March and April. Average annual rainfall, mainly registered between May and October, is about 2,400 mm, and most of the annual runoff (around 90%) occurs during that same period. Wind speed is generally low throughout the year with an average value of about 2 m/s.

2.2. Data Collection

The sampling site was located in a degraded forest, about half a kilometer from the NT2R shoreline (Lat = 17.70365°, Long = 105.16157°, see Figure 1). Ambient air gas concentrations were performed for a 2-year period (July 2010 to July 2012), when precipitation chemistry were done from June 2010 to December 2011.

2.2.1. Meteorological Conditions

Meteorological data measured directly over the NT2R water surface were obtained during four field campaigns (May 2009, March 2010, March 2011, and June 2011; Deshmukh et al., 2014). During each

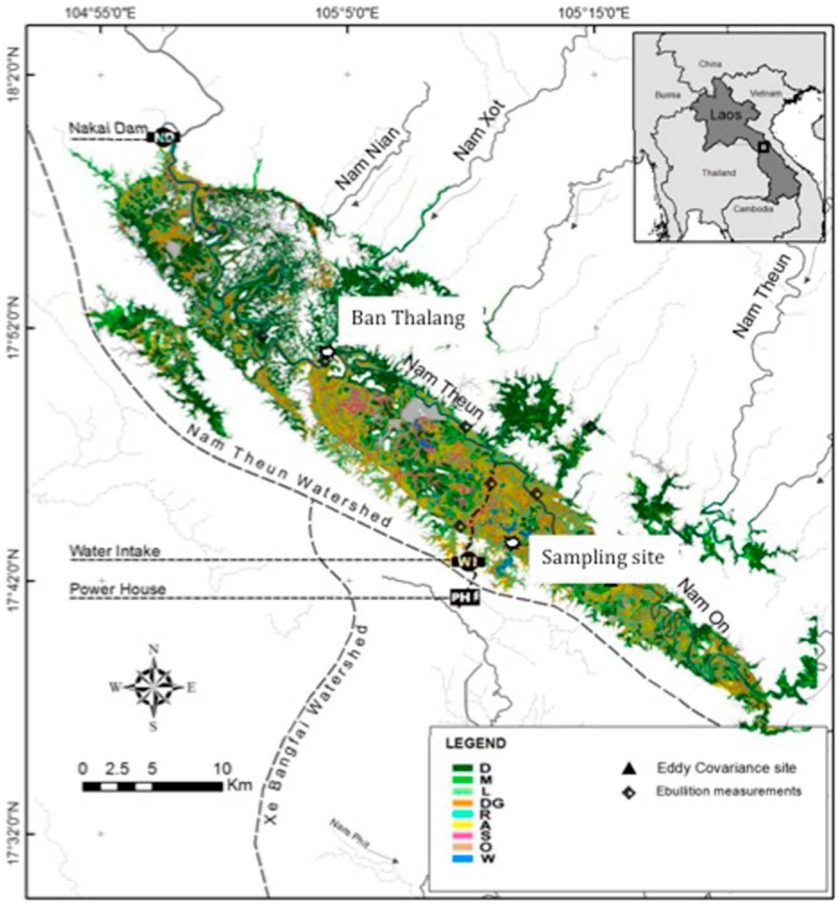

Figure 1. Contours of the NT2R at its full water level (538 m above sea level). Ecosystems present before flooding are represented: Dense [D], medium [M], light [L], degraded [DG], and riparian [R] forests; agricultural soil [A]; swamps [S]; water [W]; and *other types of surface* [O].

deployment, a meteorological station monitored continuously wind speed, atmospheric pressure, atmospheric temperature, relative humidity, and rainfall (Weather Transmitter Model WXT510, Helsinki, Finland). A radiometer (CNR-1, Kipp & Zonen, Delft, The Netherlands) was used to measure incoming shortwave radiation. The temperature of the surface water was measured 20 cm below the water surface using a thermistor (Pt 1000 sensor). All of the meteorological parameters were recorded on a CR3000 Campbell data logger. The instruments were mounted on a mast in an open water area with sensors approximately installed 3 m above the reservoir water level for each of the campaign. All the meteorological data were measured at a 1-min frequency and then processed to 3-hourly averages.

Over the study period (2004–2005, 2010–2011), precipitation data were obtained from the Ban Thalang meteorological station (Figure 1) operated by NTPC. For the years 2004 and 2005 before impoundment, a climatology of meteorological data (wind speed, atmospheric temperature, atmospheric pressure, relative humidity, rainfall rate, and solar irradiance) was obtained from the European Centre for Medium-Range Weather Forecasts (ECMWF) model. These climate reanalyzed (ERA-Interim) data sets have a 0.5° grid resolution and are provided at a 3-hr time step. More description of ERA-Interim reanalysis data can be found in Dee et al. (2011), and references therein.

2.2.2. Precipitation Chemistry

Precipitation events were collected at the sampling station using an automatic precipitation collector, which have been designed for the IDAF (IGAC/DEBITS/AFRICA-International Global Atmospheric Chemistry/Deposition of Biogeochemically Important Trace Species) network (Galy-Lacaux et al., 2009). The automatic instrument is designed to collect only wet deposition. It collects precipitation with a high degree of cleanliness in a single-use polyethylene bag, avoiding aerosol deposit before the onset of rain. A precipitation

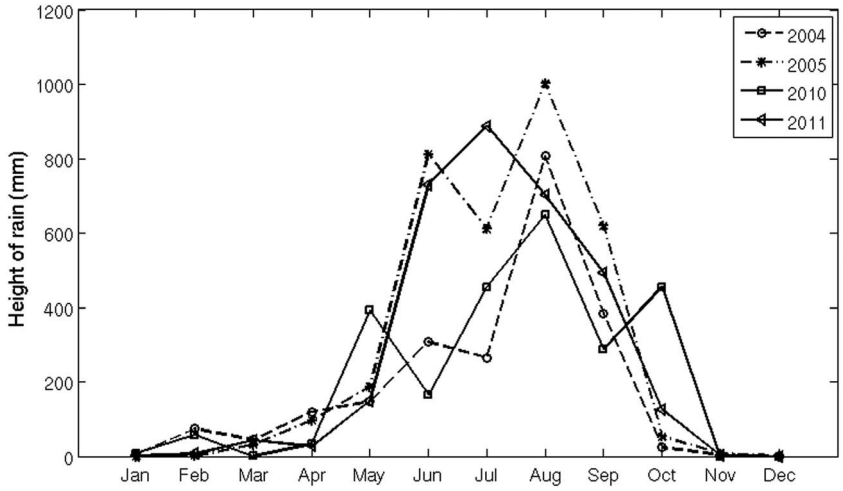

Figure 2. Monthly precipitation (mm) over the 2004–2005 (before impoundment) and 2010–2011periods (after impoundment) at Nam Theun 2 Reservoir (data provided by Nam Theun 2 Power Company).

detector automatically controls the aperture of the cover, which hermetically closes the polyethylene bag. The surface of rain collection is 225 cm^2. After each precipitation event, 50 ml of the collected precipitation are sampled in a Greiner tube and then deep freezed from collection to analysis.

Monthly precipitation data for the 2004–2005 (before impoundment) and 2010–2011 (after impoundment) periods are presented on Figure 2. Annual rainfall ranged from 2,182 to 3,420 mm with an annual mean of 2,801 mm over the 2004–2005 period, and from 2,503 to 3,162 mm (mean 2,832 mm) over the 2010–2011 period.

2.2.3. Atmospheric Concentration Measurements

Monthly atmospheric concentrations of NO_2, HNO_3, and NH_3 were measured from July 2010 to July 2012 using the passive sampling technique following the work of Ferm et al. (1994). These passive samplers were developed by the Laboratory of Aerology (LA) in Toulouse (France) in the framework of the IDAF project and tested in different tropical and subtropical regions (Carmichael et al., 2003; Ferm & Rodhe, 1997). The sampling technique is based on the property of molecular diffusion of gases and species-specific collection on impregnated filter to each pollutant measured**.** The coating solution used in the IDAF passive samplers have been specifically chosen to chemisorb targeted species into another stable species in which other pollutants do not interfere, or at least these chemical interferences are negligible. The samplers were exposed in pairs in order to ensure the reproducibility of results, and at about 10 m above the ground. The sampling procedure and chemical analysis of samples, as well as the validation method according to international standards, have been widely detailed in Adon et al. (2010). The measurement accuracy of these passive samplers, evaluated through covariance with duplicates was estimated at 9.8%, 20%, and 14.3% for NO_2, HNO_3, and NH_3, respectively. Detection limits for each trace gas were calculated from field blanks and found to be 0.07 ± 0.03 ppb for HNO_3, 0.2 ± 0.1 ppb for NO_2, and 0.7 ± 0.2 ppb for NH_3 (Adon et al., 2010).

2.3. Analytical Procedure

For precipitation, analysis set included determination of the inorganic nitrogen ions (NH_4^+, NO_3^-) mixing ratios in water by ion chromatography (IC). For gases, inorganic anions and cations, that is, nitrate, nitrite, and ammonium captured on impregnated filters of the gas passive samplers are also determined by ion chromatography. Filters are extracted in 10 ml of 18 MΩ cm water by ultrasonic stirring (15 mn). Details about analytical procedures are described by Galy-Lacaux et al. (2009) and Adon et al. (2010**)**. Basically, inorganic ions were determined with a DIONEX DX500 and ICS 1000 ion chromatograph (IC) with two automatic samplers (AS50). According to the results of the quality control intercomparison program organized twice a year by the WMO-GAW (World Meteorological Organization-Global Atmosphere Watch), analytical precision is estimated to be 5% or better for all ions, well within the uncertainties of all measured values presented here.

2.4. Estimation of Atmospheric Deposition Flux

2.4.1. Wet Deposition Calculation

Wet nitrogen deposition flux has been calculated as the product of the ammonium and nitrate concentrations in rain (monthly or annual volume-weighted mean, VWM) by the annual rainfall (Akpo et al., 2015; Galy-Lacaux et al., 2009 and references therein). Wet deposition fluxes uncertainties rely on uncertainties on rainfall amount measurements and on ionic chromatography chemical analysis. Combining these uncertainties, we estimate that the uncertainty on wet deposition fluxes measurements is about 10% (Galy-Lacaux & Delon, 2014).

2.4.2. Dry Deposition (Inferential) Modeling

Dry deposition depends both on meteorological conditions and surface characteristics. The inferential method, which combines measured air concentrations together with modeled exchange rates, was used here to estimate the dry deposition fluxes of the different gaseous species. In this study, we calculated the dry deposition velocities (V_d) of gases (NO_2, HNO_3, and NH_3) over both the NT2R water surface, and the studied area before impoundment. The V_d were calculated using the big-leaf dry deposition model of Zhang et al. (2003) that has been used in our previous study for IDAF sites (Adon et al., 2013).

The meteorological parameters were measured at about 3 m above the reservoir surface whereas the passive samplers (gas concentrations) were exposed at about 10 m above the ground. As a first approximation, using the wind profile in statically neutral conditions (Stull, 1988, equation 1), a logarithmic increase of the wind from 3 to 10 m, depending on the surface roughness, has been applied to recalculate wind speed at 10 m, and then deposition velocities at that height:

$$\overline{u}(z) = \frac{u_*}{k} \ln\left(\frac{z}{z_0}\right) \quad (1)$$

where $\overline{u}(z)$ is the mean wind speed at height z, u_* is the friction velocity, k is the von Karman constant (~0.4), z_0 is the aerodynamic roughness length.

Bidirectional exchange of NH_3 has been frequently observed over different canopies (Dorsey et al., 2004; Walker et al., 2006). Thus, to evaluate net fluxes of NH_3 over the vegetated areas (forest and rice field) present before impoundment, we used the bidirectional air-surface exchange model of Zhang, Wright, and Asman (2010), allowing for both emission and deposition computations. Note that in our study, positive fluxes are denoting deposition (and negative fluxes are denoting emission). The main parameterization of this bidirectional model, and the method to calculate the NH_3 net fluxes in this study were presented in Adon et al. (2013). The main input parameters are stomatal (Γ_{st}) and ground (Γ_g) emission potentials, leading to stomatal and soil compensation points, respectively, and thus to the canopy compensation point (X_{cp}). For each of the land use categories (LUC), Zhang, Wright, and Asman (2010) derived representative input values based on literature data. In our simulation, we used the median values of Γ_{st} (300) and Γ_g (20) suggested for tropical forest. For the rice paddies, which were weakly or not fertilized, we used the median value of Γ_{st} (631) and the lowest value of Γ_g (630) suggested for agricultural ecosystems (Zhang, Wright, & Asman, 2010) during the vegetation season, and a Γ_g of 360 for the period without vegetation (Massad et al., 2010). The average monthly simulated X_{cp} of NH_3 ranged from 0.1 to 0.3 $\mu gN/m^3$ for forest and from 1.3 to 3.6 $\mu gN/m^3$ for the rice paddies. Note that Shen et al. (2013) used X_{cp} values of 5.0 $\mu gN/m^3$ for cropland and 0.5 $\mu gN/m^3$ for forest to estimate the net fluxes of NH_3.

Before impoundment, the studied area was divided approximately into three major cover types: forest (83%), rice paddies (11%), and river surface (6% of the area). The LAI (Leaf Area Index, m^{2}/m^2) is an important parameter for calculating canopy resistances. In our study, an average LAI of 4.5 was used for the tropical forest, which is consistent with the range of values reported in Asner et al. (2003) or in Brook et al. (1999) to simulate dry deposition velocity for tropical forests. For rice paddies, LAI values were deduced from field-measured LAI during the rice-growing season (July to October) in the Jiangning County (China) reported in *Xiao* et al. (2002). The LAI increased rapidly from July to August and reached a plateau in the range of 4–5 in September.

It is important to note that few reliable methods are available to directly measure dry deposition fluxes of nitrogen species and that all estimates indirectly obtained from inferential method have a large degree of uncertainty associated to the dry deposition velocity determination. A comprehensive discussion on uncertainties related of the inferential method, based on different works (Delon et al., 2010; Flechard et al., 2011;

Table 1
Campaign Average Dry Deposition Velocities (V_d) Over the NT2R Surface (2009–2011) and the Range of Monthly Mean V_d Over the Studied Area Before Impoundment (2004–2005; see section 3.1.2)

V_d (cm/s)	NO_2	HNO_3	NH_3
NT2R surface			
20–29 May 2009	0.036 ± 0.001	0.36 ± 0.06	0.34 ± 0.07
11–25 March 2010	0.034 ± 0.002	0.37 ± 0.27	0.36 ± 0.26
12–15 March 2011	0.036 ± 0.003	0.52 ± 0.21	0.49 ± 0.23
02–08 June 2011	0.033 ± 0.001	0.28 ± 0.11	0.26 ± 0.09
Mean	0.035 ± 0.001	0.38 ± 0.10	0.36 ± 0.09
Before impoundment			
Forest	0.24–0.40	1.99–3.42	0.50–1.30
Rice fields	0.17–0.33	0.53–1.37	0.31–0.57
River surface	0.03	0.22–0.35	0.22–0.34
Mean	0.28	2.35	0.70

Zhang et al., 2009) can be found in Adon et al. (2013). Moreover, a review of biosphere-atmosphere modeling of trace gases can be found in Flechard et al. (2013), Massad and Loubet (2015), and references therein.

3. Results and Discussions

3.1. Dry Deposition Velocities

3.1.1. Over the Reservoir Surface

The mean dry deposition velocities (V_d) calculated for the NT2R surface are 0.035 cm/s, 0.38 cm/s, and 0.36 cm/s for NO_2, HNO_3, and NH_3, respectively, based on meteorological data obtained during four field measurements (Table 1). The dry deposition velocities are similar for HNO_3 and NH_3 since these gases are water soluble, that is, they deposit easily over wet surfaces such as water bodies. On the opposite, NO_2 dry deposition velocity is about 10 times smaller since this compound has a very low solubility in water. This is due to the negligible turbulent transfer processes compared to the high surface resistance for NO_2. Note that Zhang, Yu, et al. (2010) simulated an even lower NO_2 dry deposition velocity (V_d = 0.00534 cm/s) over the China seas.

For HNO_3, mean ranges of V_d from 0.28 to 0.52 cm/s over the NT2R were comparable with other estimates (0.26 to 0.42 cm/s) made over water surfaces (Matsuda et al., 2001; Table 1). As for NH_3, Schrader and Brümmer (2014) reported recent modeled deposition velocities to water (ocean) surface ranging from 0.5 to 0.9 cm/s depending on atmospheric stability and wind speeds. This range of deposition velocities over the sea is larger than our range of values (from 0.26 to 0.49 cm/s) over the NT2R. This difference could result from higher wind speeds at sea than at inland water bodies (Garland, 1977). Dry deposition velocities over the NT2R surface were modeled for the March and May–June periods to represent typical dry and wet season conditions, respectively. The seasonal V_d of HNO_3 and NH_3 are, respectively, 0.44 and 0.42 cm/s for the dry season, and 0.32 and 0.30 cm/s for the wet season. The seasonal V_d of NO_2 is 0.035 cm/s for both dry and wet seasons.

3.1.2. Over the Studied Area Before Impoundment

The ECMWF model outputs were used as meteorological inputs to simulate the dry deposition velocities over the period of 2004–2005, that is, before impoundment. The monthly mean V_d for rivers, rice paddies, and forest ranged between 0.03 and 0.40 cm/s for NO_2, between 0.22 and 3.42 cm/s for HNO_3, and between 0.22 and 1.30 cm/s for NH_3 (Table 1). The maximum values appeared in the wet season for the forested area (June–October) and for the rice paddies (September–October), while the maximum values occurred in the cold dry season (November–December) for river surface. Whatever the season, higher V_d values of HNO_3, NH_3, and NO_2 are observed in the forested area, compared to the rice paddies and water surface. This is due to the surface roughness (and atmospheric turbulence), which is the highest in forests, leading to increased dry deposition velocities in those ecosystems (Clark & Kremer, 2005; Zhang et al., 2013). Deposition velocities modeled for the forest ecosystems are in the same order of magnitude as values calculated by Adon et al. (2013) for tropical rain forest sites in Cameroon and Congo. The V_d of gases estimated for the river surface were comparable to those of the NT2R water surface after the area was flooded. The mean annual V_d before impoundment were estimated at 0.28, 2.35, and 0.70 cm/s for NO_2, HNO_3, and NH_3, respectively. In general, monthly V_d values for all species over water surface and forest simulated in this study were within a reasonable range of published values found in the literature (Adon et al., 2013; Bertolini et al., 2016; Biswas et al., 2005; Brook et al., 1999; Zhang et al., 2002, 2009, and references therein).

3.2. Dry Deposition Flux Estimates (N Species to NT2R)

3.2.1. Concentration of N Species in Air

Monthly mean concentrations of NO_2, HNO_3, and NH_3 at the monitoring station were 0.13–1.03, 0.17–0.44, and 0.33–1.67 $\mu gN/m^3$, respectively, during the study period (July 2010 to July 2012; Figure 3). On a seasonal basis, the mean concentration of NO_2 is about 2 times higher in the dry season (0.76 ± 0.05 $\mu gN/m^3$) than in the wet season (0.41 ± 0.02 $\mu gN/m^3$). On the opposite, the mean concentration of NH_3 is 2 times higher in the wet season (1.06 ± 0.06 $\mu gN/m^3$) than in the dry season (0.55 ± 0.14 $\mu gN/m^3$). Highest concentrations of NH_3

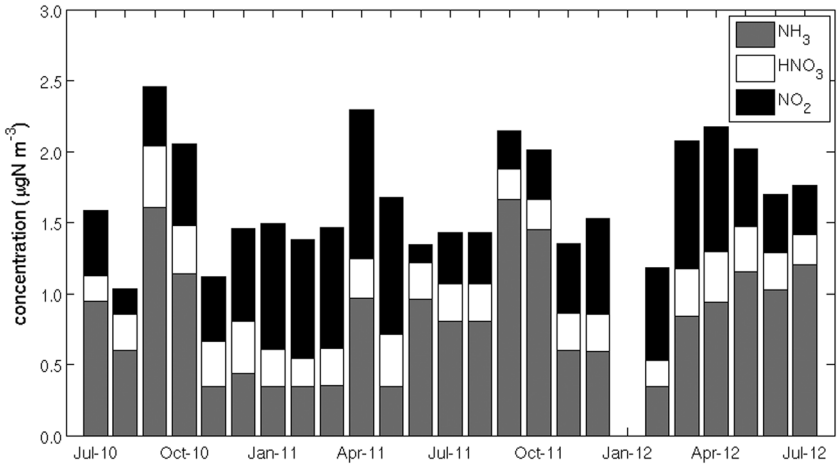

Figure 3. Monthly mean concentrations of inorganic nitrogen species in the air (NO_2, HNO_3, NH_3) at NT2R over the study period (July 2010 to July 2012). Samples were not taken in January 2012.

were observed in September and October (at the end of wet season). As for HNO_3, the dry (0.28 ± 0.01 μgN/m^3) and wet season (0.27 ± 0.02 μgN/m^3) concentrations are very comparable.

Global maps of NO_x-N and NH_3-N 2001 emissions compiled from the emission inventories of the 16 HTAP (Hemispheric Transport of Air Pollution) N oxidized models and 7 N reduced models indicated a range of 2 to 4 kgN·ha^{-1}·year^{-1} for NO_x emission and of 4 to 10 kgN·ha^{-1}·year^{-1} for NH_3 emissions in Lao PDR (Vet et al., 2014). NO_x emissions in Lao PDR are probably dominated by biomass burning especially from the frequent slash and burn agricultural practices in the country, including in the vicinity of the reservoir (Inoue et al., 2007). These emissions mostly occur during the warm dry season (March–April). Fossil fuel combustion, automobile traffic and industries seem to be of lower influence at the scale of the region and country (Inoue et al., 2007). Annual mean NO_2 concentration at the studied site (0.57 ± 0.07 μgN/m^3) is comparable to that of the forested ecosystem of Zoetele (0.52 ± 0.06 μgN/m^3), an IDAF site in Cameroon (Adon et al., 2010).

NH_3 mainly derives from the volatilizing of fertilizer and animal excrements, which are closely related with agriculture. Lao PDR is a rural country with 85% of the population depending on agriculture for their livelihood with most of the rural households producing food mainly for their own consumption. Agriculture accounts for 52% of GDP (Gross Domestic Product) with livestock and fisheries contributing 18% (Knips, 2004). In addition, the slash and burn practices could be another source of ammonia (Lobert et al., 1990). We believe that NH_3 concentrations at the studied forest site of Lao PDR represent a combination of different natural sources present at the country scale and also at the regional scale. This site is close to the Mekong central corridor, and includes contributions from cattle, biomass burning, and soil emissions (Bouwman et al., 2002; Schlesinger & Hartley, 1992). Thus, the significant contribution of soil emissions and the hydrolysis of urea from animal urine lead to higher NH_3 concentrations measured in the wet season. The annual average concentration of NH_3 observed at the studied site (0.83 ± 0.18 μgN/m^3) was lower than that measured at the forest site (2.1 ± 1.1 μgN/m^3) of the Jinjing agricultural catchment in Subtropical China (Shen et al., 2013).

3.2.2. Dry Deposition Fluxes of N Species to NT2R

Monthly dry deposition fluxes of NO_2, HNO_3, and NH_3 to NT2R were estimated from the combined monthly concentrations and seasonal dry deposition velocities of the three gases. The monthly fluxes calculated for the two sampled years have been averaged in order to give results representative of a mean climatic year. The monthly dry deposition fluxes ranged from 0.03 to 0.13 kgN·ha^{-1}·month^{-1} for NH_3, from 0.01 to 0.04 kgN·ha^{-1}·month^{-1} for HNO_3 and from 0.001 to 0.010 kgN·ha^{-1}·month^{-1} for NO_2 (Figure 4).

The relative contribution of NO_2 to total nitrogen dry deposition to the reservoir is very small in relation with its very small deposition velocity. NH_3 is the major contributor to the N dry deposition, since it cumulates both the highest concentrations and deposition velocities (Table 1 and Figure 3). We estimated higher N dry deposition fluxes in September–October at the end of the wet season due to higher NH_3 concentrations (Figure 3), and in March/April at the end of the dry season in relation to higher values of V_d.

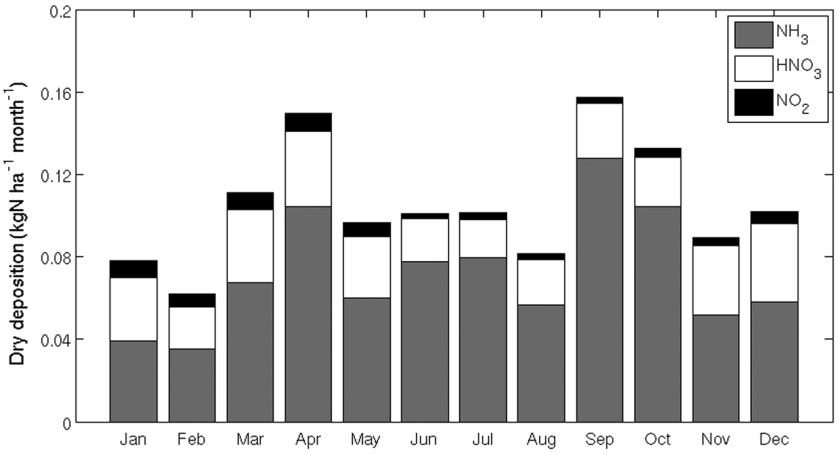

Figure 4. Monthly averaged N dry deposition fluxes of NO_2, HNO_3, and NH_3 (kgN·ha^{-1}·month^{-1}) to NT2R over the study period (July 2010 to July 2012).

On a seasonal basis, the total fluxes of NO_2, HNO_3, and NH_3 were 0.02, 0.14, and 0.51 kgN/ha in the wet season, and 0.04, 0.19, and 0.36 kgN/ha in the dry season, respectively. Total N dry deposition fluxes are of the same order in the wet season (0.67 kgN/ha) and in the dry season (0.59 kgN/ha). On an annual basis, the total dry deposition fluxes estimated were 0.06 ± 0.01 kgN·ha^{-1}·year^{1} for NO_2, 0.33 ± 0.03 kgN·ha^{-1}·year^{-1} for HNO_3 and 0.87 ± 0.18 kgN·ha^{-1}·year^{-1} for NH_3 or a total annual N dry deposition of 1.26 ± 0.14 kgN·ha^{-1}·year^{-1} over the study period. The contributions of NH_3 and HNO_3 to the total N dry flux were 68% and 27%, respectively.

Dry deposition fluxes over reservoirs or water bodies are scarce in the literature, particularly for gaseous species. Indeed, studies on nitrogen deposition to water surface (rivers, bays, and lakes) have rather focused on dry deposition of aerosol particles (Chen et al., 2011; Gao, 2002; Jung et al., 2009). For example, Ayars and Gao (2007) estimated a total particulate N (pNH_4^+ + pNO_3^-) dry deposition of 1.54 kgN·ha^{-1}·year^{-1} over the Mullica River-Great Bay Estuary. Other studies included dry deposition for both gaseous and particulate forms over lake, seas, and oceans (Luo et al., 2014; Zhan et al., 2017; Zhang et al., 2013; Zhang, Yu, et al., 2010). Therefore, it is difficult to compare theirs estimates to our result in this study. Nevertheless, our estimate for the NT2R (1.26 kgN·ha^{-1}·year^{-1}) lie within the range value of total dry deposition fluxes (0.5–5 kgN·ha^{-1}·year) over the Eastern China Seas estimated by Zhang, Yu, et al. (2010). For the Lake Tahoe water surface in California, Tarnay et al. (2001) estimated the total N dry deposition (including particulate N) to be in the range of 0.57–1.2 kgN·ha^{-1}·year^{-1} for the summer and dry fall season. Our estimate for the NT2R is comparable to the previous value, but it should be noted that our study does not include N dry deposition fluxes for nitrogenous particles.

3.3. Wet Deposition of N Species

3.3.1. Concentration of N Species in Rainwater

The highest nitrogenous compounds concentrations were measured in the first rainy events sampled between February and April, in the period still defined as the dry season (Figure 5). N concentrations varied from 0.32 to 1.34 mgN/L for NO_3^-, and from 0.41 to 1.88 mgN/L for NH_4^+.

In the wet season (May to October), measured inorganic nitrogen VWM concentrations were much lower, between 0.01 and 0.17 mgN/L for NO_3^- and between 0.02 and 0.23 for NH_4^+. In the dry season, due to the much lower frequency of rainfall events and subsequent washout/rainout, N atmospheric content (particles and gases) was highest and maximum nitrate and ammonium concentrations in precipitation were measured. On the opposite, the higher frequency and amount of rainfall during the wet season led to low concentrations of nitrogen in rain. Annual VWM concentration of NO_3^- measured in 2011 was 0.06 mgN/L, and 0.10 mgN/L for NH_4^+. The year 2010 was not completely sampled and excluded the first rainy month of May. However, annual VWM concentrations of NO_3^- and NH_4^+ with 0.05 and 0.09 mgN/L′ respectively, are comparable to the ones found for 2011. That incompletely sampled year certainly represents the lower

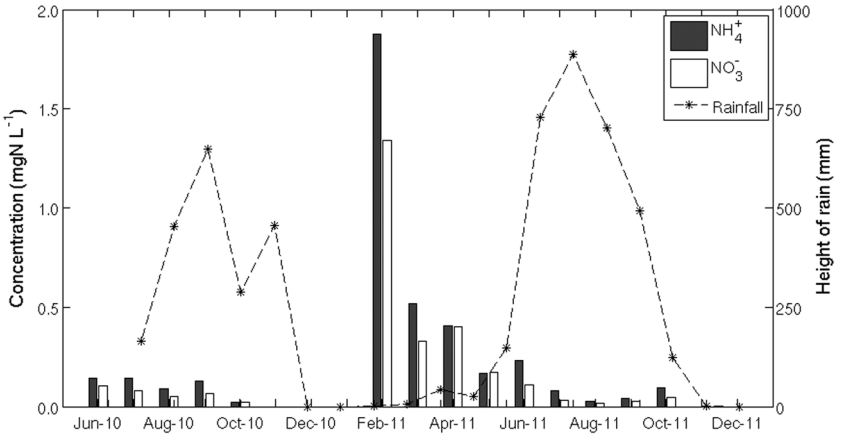

Figure 5. Monthly mean concentrations (in mgN/L) of inorganic nitrogen species in rainwater (NH_4^+, NO_3^-) at NT2R from June 2010 to December 2011.

range of annual VWM nitrate and ammonium concentrations for that site. In 2011, sampling is representative for all months of the year. Total annual concentration of nitrogen measured at the NT2R ranged from 0.14 to 0.16 mgN/L for the 2 years. If we consider the rural or coastal sites mentioned in the last WMO assessment on deposition (Vet et al., 2014), our results fall in the lower part of the reported range of N concentration in rain as compared to measurements made in Thailand, Vietnam, India, and South/Southeast China (0.05 to 0.8 mgN/L).

3.3.2. Wet Deposition Fluxes

The monthly wet deposition fluxes for rainy months ranged from 0.09 to 1.67 kgN·ha^{-1}·month^{-1} for NH_4^+–N and from 0.06 to 0.81 kgN·ha^{-1}·month^{-1} for NO_3^-–N (Figure 6). The highest fluxes for both NH_4^+–N and NO_3^-–N occurred in the wet months of June, July, and August. On a seasonal basis of the period 2010–2011, wet deposition flux equaled 2.67 kgN/ha for NH_4^+–N and 1.54 kgN/ha for NO_3^-–N in the wet season, or a total of 4.21 kgN/ha. In the dry season (November to April), which gathers only 2% to 4% of the total annual rainfall during the years 2010–2011, wet deposition fluxes were estimated to be 0.46 kgN/ha for NH_4^+–N and 0.34 kgN/ha for NO_3^-–N, or a total of 0.8 kgN/ha.

At the annual scale, wet deposition fluxes of NH_4^+–N and NO_3^-–N were 3.13 ± 0.71 and 1.88 ± 0.21 kgN·ha^{-1}·year^{-1}, respectively, or a total inorganic N wet deposition of 5.01 ± 0.92 kgN·ha^{-1}·year^{-1} over the period 2010–2011. Total annual nitrogen wet deposition occurred at 62.5% in the reduced form (NH_4^+) and 37.5% in the oxidized one (NO_3^-). Mean annual N wet deposition fluxes reported by Vet et al. (2014)

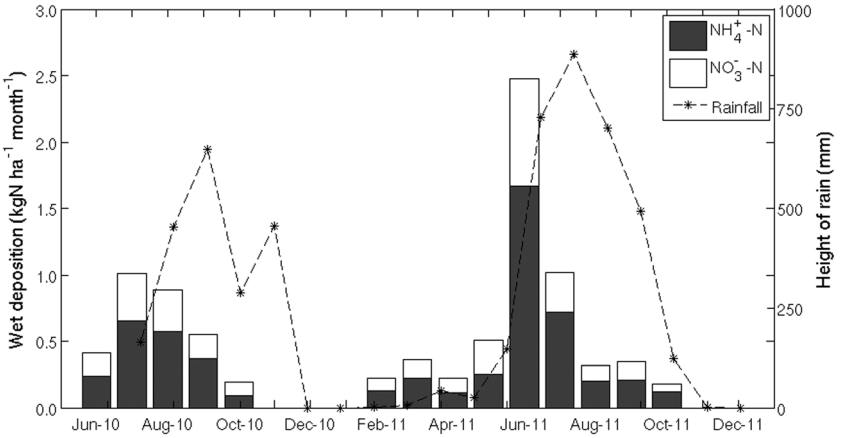

Figure 6. Monthly nitrogen wet deposition fluxes of NH_4^+–N and NO_3^-–N (kgN·ha^{-1}·month^{-1}), and precipitation (height of rain, mm) to NT2R from June 2010 to December 2011.

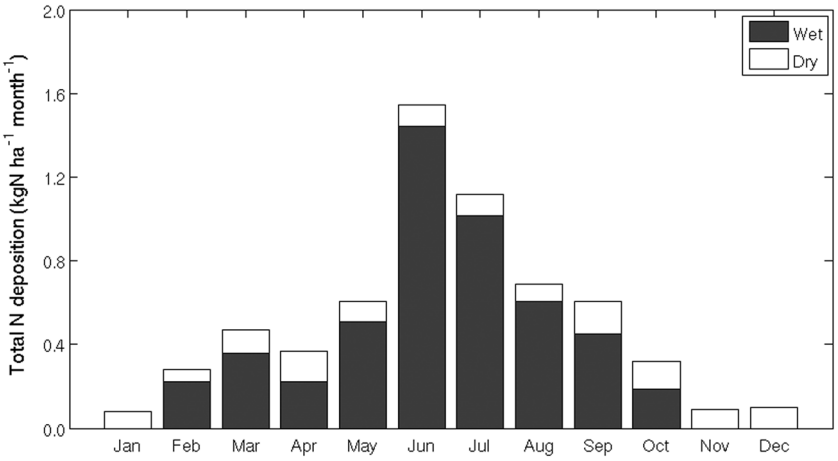

Figure 7. Monthly total (dry + wet) N deposition fluxes (kgN·ha^{-1}·month^{-1}) to NT2R.

ranged between 4 and 8 kgN·ha^{-1}·year^{-1} for different sites in India, Thailand, and Vietnam for the period from 2000 to 2002, a range compatible with data reported here.

3.4. Total Nitrogen Deposition Fluxes to NT2R

For each deposition process (dry or wet), monthly fluxes were averaged for the two years. The monthly total (wet + dry) atmospheric N deposition fluxes ranged from 0.08 to 1.55 kgN·ha^{-1}·month^{-1} (Figure 7).

Total N deposition appeared to be highest in the wet season (May through October), with a wet season total flux of 4.88 kgN/ha. The dry season (November to March) total N deposition flux was estimated to be 1.39 kgN/ha. On an annual basis, the total N deposition flux directly to the NT2R surface was 6.27 ± 1.06 kgN·ha^{-1}·year^{-1} over the study period. Wet deposition is the dominant removal process, accounting for 80% of the total N flux. The reduced (NH_4^+, NH_3) and oxidized (NO_3^-, HNO_3, NO_2) forms of the total N deposition flux were calculated to be 3.99 kgN·ha^{-1}·year^{-1} (63.6%) and 2.28 kgN·ha^{-1}·year^{-1} (36.4%), respectively.

In comparison to other water surfaces (Table 2), the magnitude of the total N deposition flux to NT2R is similar to the deposition on the coastal Barnegat Bay, New Jersey (Gao, 2002), but lower than on the Great Bay Estuary, New Jersey (Ayars & Gao, 2007). Jung et al. (2009) reported also a higher value of 16.6

Table 2
Comparison of Total N (Dry + Wet) Deposition to Water Surface With Other Studies

			Wet deposition	Dry deposition	Total N deposition	
	Collection year	Nitrogen species	Flux (kgN·ha^{-1}·year^{-1})	Flux (kgN·ha^{-1}·year^{-1})	Flux (kgN·ha^{-1}·year^{-1})	References
NT2 Reservoir, Laos	Jun 2010 to Jul 2012	NH_4^+, NO_3^-, NH_3, HNO_3, NO_2	5.01	1.26	6.27	This study
Lake Sihwa, South Korea	2004	NH_4^+, NO_3^-, pNH_4^+, pNO_3^-	10.3	6.30	16.6	Jung et al. (2009)
River-Great Bay Estuary, New Jersey, USA	Mar 2004 to Mar 2005	NH_4^+, NO_3^-, pNH_4^+, pNO_3^-	9.98	1.54	11.52	Ayars and Gao (2007)
Barnegat Bay, New Jersey, USA	Feb 1999 to Feb 2001	NH_4^+, NO_3^-, pNH_4^+, pNO_3^-	6.69	0.77	7.46	Gao (2002)
Lake Tahoe, California-Nevada	Jul–Sep 1997 and Jul–Sep 1998	NH_4^+, NO_3^-, NH_3, HNO_3, pNH_4^+, pNO_3^-	1.7–2.9	0.57–1.2	2.27–4.1	Tarnay et al. (2001)
Lake Dianchi, China	Apr 2010 to Mar 2011	NH_4^+, NO_3^-, NH_3, NO_2, HNO_2/HNO_3, pNH_4^+, pNO_3^-	10.9	11.17	22.07	Zhan et al. (2017)

kgN·ha^{-1}·year^{-1} for the total annual deposition of N to Lake Shiwa under the influence of high anthropogenic emissions of NO_x and NH_3 from urban sources or long-range transport of N species. Over Lake Dianchi in China, Zhan et al. (2017) estimated the highest value of N dry deposition flux (11.17 kgN·ha^{-1}·year^{-1}) with a contribution of 94% for gaseous N species (10.5 ± 0.3 kgN·ha^{-1}·year^{-1}) and 6% for particulate inorganic N species (0.67 ± 0.1 kgN·ha^{-1}·year^{-1}). Dry deposition of gaseous N species over NT2R in our study (1.26 ± 0.14 kgN·ha^{-1}·year$_{)}^{-1}$ is 88% lower than that reported for the Lake Dianchi.

It is worth mentioning that dissolved organic nitrogen (DON) could be an important portion of N in precipitation (Cornell et al., 2003; Zhan et al., 2017). As a matter of consequences, the results from this study need to be considered as the lower estimate value for the total atmospheric N deposition since neither DON, nor particulate nitrogen (pNH_4^+ and pNO_3^-) were estimated here. Nevertheless, we tried to give a rough estimation of particulate dry deposition in our study. Compared with gaseous N species, particulate N species were not the major contributor of dry-deposited N; they contributed approximately 6–10% to the total flux (Pan et al., 2012; Zhan et al., 2017). Supposing a contribution of 10% for particulate species to the total flux in our study, dry deposition of particulate inorganic N estimated would be 0.14 kgN·ha^{-1}·year^{-1}, leading to a total (gaseous and particulate) N dry flux of 1.4 kgN ha^{-1} yr^{-1} deposited at NT2R. Accordingly, the total (dry + wet) N deposition flux estimated would be 6.41 kgN·ha^{-1}·year^{-1} over NT2R with 78% from wet deposition. Considering the value (including the standard deviation) of gaseous N dry deposition flux (1.26 ± 0.14 kgN·ha^{-1}·year^{-1}) over the study period, we note that the underestimation of N budget is not so significant if particulate N dry deposition was not taken into account.

3.5. N Deposition Change Due to Reservoir Flooding

3.5.1. Before Impoundment

We estimated nitrogen deposition flux in the studied area before impoundment in order to quantify the net change in atmospheric deposition resulting from the reservoir impoundment. As the sampling of gases and precipitation chemistry started on 2010, after the impoundment in 2008, we have made some assumptions for the post impoundment conditions. These hypotheses were made on the sources of atmospheric gases and aerosols concentrations and on subsequent total N deposition fluxes estimation that we can make before impoundment. Since reactive N emissions (NO_x and NH_3) from soils are higher compared to aquatic systems, as a first approximation, we assumed that local N atmospheric gases concentrations over the flooded area might be higher before impoundment than after impoundment. On the other hand, atmospheric deposition can be a significant source of nitrogen in terrestrial ecosystems (Law, 2013) compared to aquatic ecosystems. As a matter of consequences, and without further precisions on the pre-impoundment conditions, we assumed that the sources of gases and aerosols may have changed within a ±30% range when compared to the measured postimpoundment conditions. As a matter of consequence, we applied a range of ±30% to the gaseous concentrations used to estimate dry deposition fluxes before the creation of the reservoir. Furthermore, for dry deposition, the meteorological conditions and surface characteristics of the ecosystem have been taken into account in the simulation of dry deposition velocities for the different land covers (forest, rice paddies, and river surface) before impoundment (section 3.1.2). By applying the ±30% variability to the monthly N concentrations measured in air, the range of mean annual concentrations were estimated to be 0.41 to 0.77 μgN/m^3 for NO_2, 0.56 to 1.04 μgN/m^3 for NH_3, and 0.20 to 0.36 μgN/m^3 for HNO_3 before impoundment. Monthly mean dry deposition fluxes were estimated using two scenarios: N concentrations were supposed to be 30% higher before impoundment (high scenario), and N concentrations were supposed to be 30% lower before impoundment (low scenario) than after impoundment. Only annual mean dry deposition fluxes are discussed here.

For the high scenario, on an annual basis, the dry deposition fluxes of HNO_3 and NO_2 to the forest were estimated to be, respectively, 3.06 and 0.69 kgN·ha^{-1}·year^{-1}. The net dry deposition flux of NH_3 to the forest was estimated at 2.04 kgN·ha^{-1}·year^{-1}. On this basis, for the forest, the total N ($HNO_3 + NO_2 + NH_3$) dry deposition was estimated to be 5.79 kgN·ha^{-1}·year^{-1}. For the rice paddies, the mean annual dry deposition fluxes of HNO_3 and NO_2 to the rice field were estimated to be, respectively, 0.99 kgN·ha^{-1}·year^{-1} and 0.52 kgN·ha^{-1}·year^{-1}. The net emission flux of NH_3 from the rice paddies was estimated to be −2.81 kgN·ha^{-1}·year^{-1}. Then, the total annual N net emission flux was estimated to be −1.30 kgN·ha^{-1}·year^{-1} for the rice paddies. For the water surface, the total annual N dry deposition flux was estimated to be 1.31 kgN·ha^{-1}·year^{-1}. Calculating the land use weighted dry N deposition, it comes

that the total N dry deposition to the studied area was estimated to be 4.75 kgN·ha^{-1}·year^{-1} before impoundment. Total N dry deposition in the studied area is mainly driven by the N deposition in forest, since the forest is the major land cover in the studied area (83%) associated with the highest deposition velocity. For the low scenario, total N dry deposition flux was estimated at 2.82 kgN·ha^{-1}·year^{-1} on the forested ecosystem and 0.71 kgN·ha^{-1}·year^{-1} on the water surface. For the rice paddies, total N net flux was estimated at −2.67 kgN·ha^{-1}·year^{-1} due to the high net NH_3 emission (−3.5 kgN·ha^{-1}·year^{-1}) compared to deposition fluxes of NO_2 (0.28 kgN·ha^{-1}·year^{-1}) and HNO_3 (0.53 kgN·ha^{-1}·year^{-1}). The estimated land use weighted dry N deposition at the studied area was 2.09 kgN·ha^{-1}/year^{-1} before impoundment. By combining the two scenarios, the total N dry deposition to the studied area ranged from 2.09 to 4.75 kgN·ha^{-1}·year^{-1} before impoundment, an average of 3.42 ±1.88 kgN·ha^{-1}·year^{-1}.

Wet deposition fluxes are directly linked to the annual rainfall that varied from 2,182 to 3,420 mm/year with an interannual variability of 22% over the period 2004–2007 before impoundment and from 2,165 to 3,162 mm/year with an interannual variability of 15% over the period 2008–2012 after impoundment. Knowing that estimated uncertainty on wet deposition fluxes measurement is about 10% (Galy-Lacaux & Delon, 2014), it seems reasonable to consider a total variability of ±30% for N wet deposition fluxes. That variability results from the natural variability of rainfall and integrated the variability of the different atmospheric sources of gases and particles influencing nitrogen rain content. This percentage of variability is close to the ±25% maximum variation for total N wet deposition fluxes found by Galy-Lacaux and Delon (2014) based on a synthesis on the different African IDAF sites (dry savanna, wet savanna, and forest). The amount of N wet deposition on the NT2R was estimated to be 5.01 ± 0.92 kgN·ha^{-1}·year^{-1} after the impoundment. By applying the ±30% variability to that wet deposition flux, it comes a range from 3.51 to 6.51 kgN·ha^{-1}·year^{-1} for the wet deposition before impoundment, an average of 5.01 ±2.12 kgN·ha^{-1}·year^{-1}. Vet et al. (2014) reported a comparable range of 4 to 8 kgN·ha^{-1}·year^{-1} for mean annual N wet deposition fluxes in different sites in Thailand and India for the period 2000–2002.

Combining both dry and wet deposition fluxes and considering the two scenarios, the total N deposition flux ranged from 5.60 to 11.26 kgN·ha^{-1}·year^{-1} over the studied area before impoundment corresponding to an average of 8.43 ±4.01 kgN·ha^{-1}·year^{-1}. Dry deposition process in gaseous form contributes 37–42% of the total N flux, that is, dry deposition is almost as important as wet deposition for direct N input to the ecosystem.

Shen et al. (2014) estimated much higher total N deposition of 46.3 kgN·ha^{-1}·year^{-1} at the agricultural catchment of Jinjing in subtropical central China. The Jinjing catchment has forest, rice paddies, and tea plantations as the main land use types, accounting for 65.5%, 26.5%, and 2.4% of the total catchment area, respectively. The other minor land uses in the catchment include reservoir, river, and others. In addition to nitrogen species considered in our study, their estimate included DON and nitrogen particulate (pNH_4^+ and pNO_3^-). By considering the same N species as in our study (i.e., without DON and nitrogen particulate), their estimate was 32.1 kgN·ha^{-1}·year^{-1}, still around 4 times higher than our estimate. This difference is mainly due to high concentrations of N compounds in both the rainwater and the ambient air measured in the Jinjing agricultural catchment (Shen et al., 2013). This result can be considered as an indication that the studied site of Lao was probably under minor anthropogenic influence as opposed to subtropical central China where heavy fertilization is common.

3.5.2. Impact of the Flooding

When considering the mean of the range of the total N deposition flux before impoundment (8.43 ±4.01 kgN·ha^{-1}·year^{-1}), the total N deposition flux after impoundment to the water surface of NT2R (6.27±1.06 kgN·ha^{-1}·year^{-1}) would represent a 26% decrease of N deposition. We found that this difference is mainly due to a reduction of 63% of N dry deposition from the vegetated surface (3.42 kgN·ha^{-1}·year^{-1}) present before the reservoir impoundment to the water surface of NT2R (1.26 ± 0.14 kgN·ha^{-1} year^{-1}). Note that if we considered the higher end of the range, the total N deposition flux before impoundment (11.26 kgN·ha^{-1}·year^{-1}) would be reduced by 44% after impoundment (with a reduction of 73% for N dry deposition). The N wet deposition flux is mainly influenced by the amount of the precipitation and its variability, both parameters that are not related to the impoundment. Indeed, no change in measurements of precipitation pattern (amount and seasonality) in relation with the impoundment has been observed at the studied area.

It should be noted that the total N deposition estimated in this study for the ecosystem may have some limits. First, we supposed a 30% variability of wet deposition fluxes and of N concentrations measured in the air after impoundment to estimate a likely range of N dry deposition flux before impoundment. In addition, we hypothesized that the concentrations measured at the forest site would apply in rice paddies and river surface, although Shen et al. (2013) measured higher NH_3 concentrations in the rice paddies (3.2 $\mu gN/m^3$) than in the forest site (2.1 $\mu gN/m^3$), and conversely lower NO_2 concentrations in the rice paddies (4.1 $\mu gN/m^3$), compared to the forest site (6.1 $\mu gN/m^3$). Note that the higher concentrations of NH_3 in the rice paddies of Jinjing catchment was most likely due to the intense use of N fertilizer (360 $kgN{\cdot}ha^{-1}{\cdot}year^{-1}$) in those fields (Shen et al., 2013). We note that the use of fertilizer in Lao PDR is the lowest in Asia (FAO report, 2011) and NH_3 concentrations at Lao forest site (0.83 $\mu gN/m^3$) represent a combination of low-intensity sources at the regional scale. We assumed that these assumptions have not such an impact, since the forest was the major land use of the studied area and the sampling site was located in a forest close to the NT2R.

For the first time, we estimated the change in the nitrogen deposition budget resulting from a reservoir impoundment. This result indicates a decrease of N deposition flux mainly due to the decrease of the dry deposition component. This can be expected on a water body as a consequence of the reduction of the dry deposition velocities on water surface when compared to preexisting vegetation. A considerable amount of research has been done on the effects of N deposition to terrestrial and aquatic ecosystems (Galloway et al., 2008, and references therein). For example, N deposition can degrade N retention for whole forest ecosystems leading to eutrophication in adjacent aquatic ecosystems (Stoddard, 1991). However, no study was conducted on the impact of atmospheric N deposition following water impoundment and more research is necessary to evaluate this impact on atmospheric chemistry.

4. Conclusions

This study provides an estimation of the atmospheric nitrogen deposition over the NT2 hydroelectric reservoir and on the area that was flooded before the reservoir creation. The total (dry + wet) annual nitrogen deposition flux at the reservoir surface is estimated to be 6.27 ±1.06 $kgN^{-1}ha^{-1-1}year^{-1}$. Wet deposition was the major source of the atmospheric nitrogen input to the NT2R, contributing to around 80% of the total atmospheric deposition. More than 84% of the annual wet N deposition (5.01 ± 0.92 $kgN^{-1}ha^{-1-1}year^{-1}$) occurred during the wet season (May through October), along with 96–98% of the recorded annual rainfall. Ammonium was the largest contributor (62.5%) to the total N wet deposition, compared to nitrate. Dry N deposition flux, calculated from NH_3, HNO_3, and NO_2 concentration measurements, is estimated at 1.26 ± 0.14 $kgN^{-1}ha^{-1-1}year^{-1}$ on the NT2R water surface. The mean total nitrogen deposition flux has been also estimated to be 8.43 ± 4.01 $kgN^{-1}ha^{-1-1}year^{-1}$ over the studied area dominated by forests before impoundment. Total N deposition over the studied area was reduced of 26% after impoundment based on our working hypothesis (application of a variability of 30% to N concentrations in air and to N wet deposition flux before impoundment). The dissolved organic nitrogen and particulate nitrogen have not been taken into account in this budget. Therefore, our estimates of atmospheric N deposition should be regarded as a lower limit of nitrogen deposited from the atmosphere.

Acknowledgments
The authors are grateful to the Nam Theun 2 Power Company and the Government of Laos (Lao PDR) for the various permissions and authorizations granted as well as their help with logistics during field campaigns. We would like to thank Phonesavanh Xayyavong for collecting precipitation and gas data during the sampling period. The data used in this study are original and are publicly available in Tables S1–S5 in the supporting information.

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