

Inorganic nitrogen dynamics in the River Seine downstream from Paris (France)

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Abstract. The River Seine, below Paris, receives the effluents from a large sewage treatment plant, increasing the ammonium concentration up to 6 mgN.l⁻¹ in late summer. Careful measurement of ammonium, nitrate and organic nitrogen during the downriver travel of the water masses over 100 km below the outfall, along with direct determination of nitrification and benthic fluxes, allowed to establish a budget of nitrogen transport and transformations in this reach of the river. Nitrification is shown to start after a distinct period of several days required for the growth of a significant nitrifying bacterial population. Denitrification is active in the upper layer of bottom sediments but absent from the water column. Comparison of our data with those published for the period 1973–1976 shows that the nitrate load carried by the river has increased not only because of higher runoff of agricultural nitrate in the upstream part of the watershed, but also as a result of the severe reduction in the rate of denitrification processes, owing to the restoration of better oxygen conditions.

Introduction

Riverine transport of nitrogen from land to the coastal marine environment is a process of great significance at the local as well as at the global scale. Its increase as a result of human activities is the cause of severe problems of coastal eutrophication (Lancelot et al. 1987) and, in the long term, might alter the global circulation of biogenic elements.

Rivers draining forested or agricultural watersheds receive nitrogen primarily under the form of nitrate, easily leached from soils, or organic nitrogen, from the biota. When draining urbanized areas, they often receive in addition high amounts of ammonium, which is the major form of nitrogen in the outflow of standard (activated sludge) purification plants. Ammonification of organic nitrogen, nitrification and denitrification may considerably affect the behaviour of nitrogen during its downriver transfer.

Denitrification is especially important because it results in the elimination of nitrogen from the system and thus can significantly modify the total amount ultimately discharged into the sea (Billen 1990). This process occurs either in the sediments or in the water column when anoxic conditions are established. It is therefore particularly intense in large, highly polluted rivers, with limited reaeration and important mud deposits.

Nitrification, on the other hand, is an aerobic process, converting ammonium into nitrates. The large potential for nitrification in small rivers (with hydraulic radius lower than about 1–5 m) receiving large inputs of ammonium has long been recognized (Gujer 1976; Cooper 1984). It was also clearly demonstrated that nitrification in these systems is mostly associated with the benthos (Schwert & White 1974; Cooper 1984). The slow growth rate of nitrifying bacteria, on the one hand, and their tendency to attach to solid surfaces, on the other hand, probably explains that very few are found in the free water phase of small rivers. The situation is quite different in large estuaries, however. The much lower bottom area per unit water volume and the fact that sediments are often anaerobic within a few millimeter of the surface strongly limit the significance of benthos as a site for nitrification of water column ammonium. The presence of high concentrations of suspended matter with longer residence time than the water itself (turbidity maximum) often offers a site for attachment of nitrifiers and leads to enhanced water column nitrification in estuaries (Billen 1975; Somville 1978; Helder et al. 1983; Owens 1986; Berounsky & Nixon 1990). The situation in large rivers is intermediate. For the same reasons as in estuaries, the contribution of the benthos to nitrification is rather limited. How far these rivers are able to oxidize large inputs of ammonium will therefore depend on the ability of nitrifying bacteria to develop significant populations in the water column. Admiraal et al. (1989) recently examined this question in the lower river Rhine, using data on the decrease in ammonium concentration over three different branches of the river. They concluded that factors leading to resuspension of sediments, like high flow rate of water or turbulence induced by intense shipping are major determinants of nitrification rates there.

The river Seine below Paris also offers an interesting system for examining the role of nitrification and denitrification (both in the water column and in the benthic phase) on the dynamics of inorganic nitrogen in a perturbed lotic environment.

Indeed, the discharge of the large sewage treatment plant of Achères, located at about 40 km below Paris and treating most of the wastewater from this large city, results in a sudden and tremendous increase in ammonium concentration. The river then flows for more than 100 km

without any further significant perturbation, until the beginning of the estuarine zone. In this paper, we present data collected on ammonium and nitrate concentration, abundance and activity of nitrifying bacteria and benthic denitrification in the lower Seine, and discuss their significance in the balance of nitrogen transport to the estuarine zone.

Biotope and methods

Studied area

The river Seine, below Paris is a large 7th order, regulated river, draining about 44000 km². The median discharge at Paris is 156 m³/s and ranges from about 30 m³/s in dry summer to 1820 m³/s under flood conditions, generally observed in winter. 60 km below Paris, it receives the river Oise, draining a watershed of 16980 km², with a mean discharge of 75 m³/s (range 25–535 m³/s). The Seine river becomes of 8th order after this confluence. The stretch of the river extending downstream from the Oise confluence to Poses, located at the upper limit of the tidal influence, consists in a succession of reaches separated by lock-gates, maintaining a mean depth of about 5 m (mean wetted section 800 m²).

The river receives all wastewater from the Paris agglomeration (10 million inhabitants). About 80% of the wastewater reaches the site of Achères, located 8 km upstream the Oise confluence. Since 1978, the purification plant of Achères is able to treat about 70% of the wastewater by the conventional activated sludge process, without nitrification. The remaining 30% is discharged into the river without treatment, either at Achères or upstream within the Parisian suburbs.

Sampling

Some conclusions reached in this paper rest on careful sampling procedures adopted in order (i) to obtain representative samples in spite of possible transversal heterogeneity of the river, and (ii) to follow a given water mass during its downriver travel.

(i) Problems of transversal (and vertical) heterogeneity are particularly severe in the vicinity of the outfall from the treatment plant and the Oise confluence (Chesterikoff et al. 1991). In these areas, water was pumped from 2 depths (0.5 and 4 m) at 3 points of the cross-section of the river, and mixed for providing a representative sample. Elsewhere, the samples were made by mixing water pumped from two depths in the middle of the river.

(ii) To identify a given water mass during its downriver pathway, we continuously recorded the variations of conductivity at several stations along the river. These variations result from the strong daily variations of wastewater discharges at Achères. They make up a natural signal the transmission of which could be easily followed down to Poses, although considerably damped (Chesterikoff et al. 1991). Using this signal as a marker of a given water mass at the level of Achères, it was possible to follow it during the time required for getting to Poses (about one week by low flow).

We also made use of nitrogen concentration data obtained in synchronized profiles sampled monthly by the Water Agency of the Seine-Normandy Basin (AFBSN) and published as Water Quality Yearbook. For these data, the sampling precautions described above were not used.

Analytical methods

Samples for chemical analysis were filtered through glass fiber filters and kept frozen until analysis.

Nitrate (+ nitrite) was determined by spectrophotometry after reduction

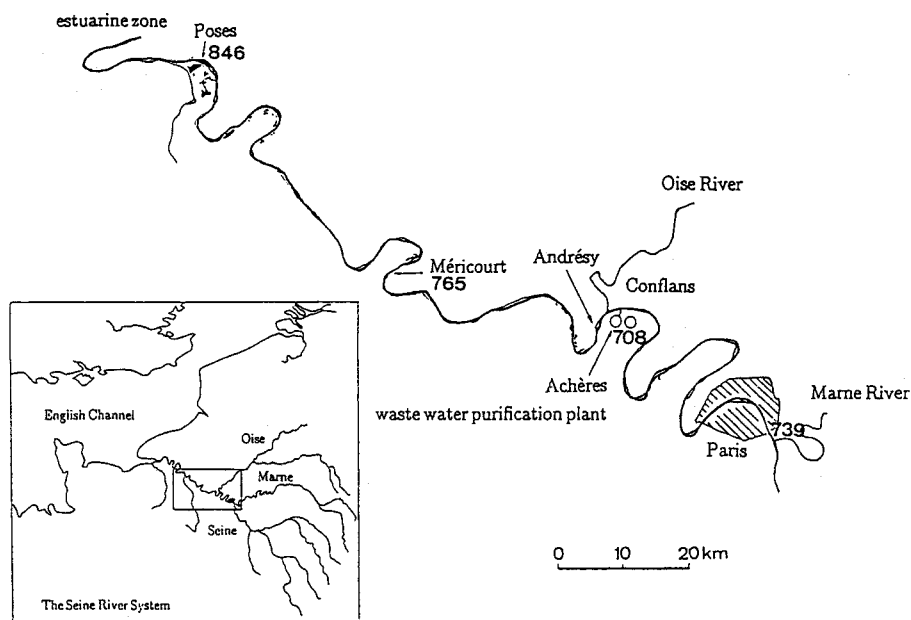


Fig. 1. Map of the lower Seine River. The stations are located by a kilometric unit, pK, used by the Financial Agency of the Seine–Normandy Basin. pK is set equal to 1000 at Honfleur, the mouth of the estuary and then decreases upwards.

with cadmium as described by Jones (1974). Ammonium concentration was measured by indophenol blue colorimetry (AFNOR 1979). Total nitrogen was determined as nitrate after alkaline persulfate mineralization (Solorzano & Sharp 1980). Organic nitrogen was estimated by difference.

Microbiological measurements

Abundance of nitrifying bacteria was evaluated by the most probably number (MPN) method. 10 samples of 0.1 ml, 10 of 0.01 and 10 of 0.001 ml were inoculated under sterile conditions in about 5 ml of the following enrichment medium (Rodina 1972):

$(\text{NH}_4)_2\text{SO}_4$	1.32 g.l ⁻¹
K_2HPO_4	1
$\text{MgSO}_4 \cdot 7\text{H}_2\text{O}$	0.5
$\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$	0.3
CaCO_3	precipitated
NaCl	2
distilled water	1000 ml

One ml of a solution of chelated metals (Carlucci & Strickland 1968) was added to the above medium and the pH adjusted to 7.5 with NaOH. Production of nitrates was qualitatively detected with nitrate-sensitive paper (*Merckoquant Nitrate Test*).

Nitrifying activity was determined according to the N-serve inhibition/¹⁴C-bicarbonate incorporation method described by Billen (1976) and adapted by Somville (1978) for river water.

Flux across the sediment water interface were determined using a 10 liter bell-jar with an open section of 855 cm², equipped with a stirring system, deposited on the bottom. Water samples from inside the bell were pumped with a syringe through a long silicone rubber capillary at 1–3 hours intervals. Oxygen concentration was continuously recorded by a YSI oxygen probe. Flux rates were calculated on the basis of the observed variations of concentration inside the bell-jar during the first 1–4 hours.

Results

Relationships between NH_4 and NO_3 concentration and river flow

Values of NH_4 and NO_3 concentrations observed at either Méricourt (pK 765) or Porcheville (pK 749) over the period 1983–1990 are plotted

against river discharge in Fig. 2a and 2b respectively. NH_4 (mgN/l) concentration obeys the following decreasing hyperbolic relation with discharge (Q , m^3/s) (Fig. 2a):

$$\text{NH}_4 = (825/Q) - 0.175 \quad (r = 0.93)$$

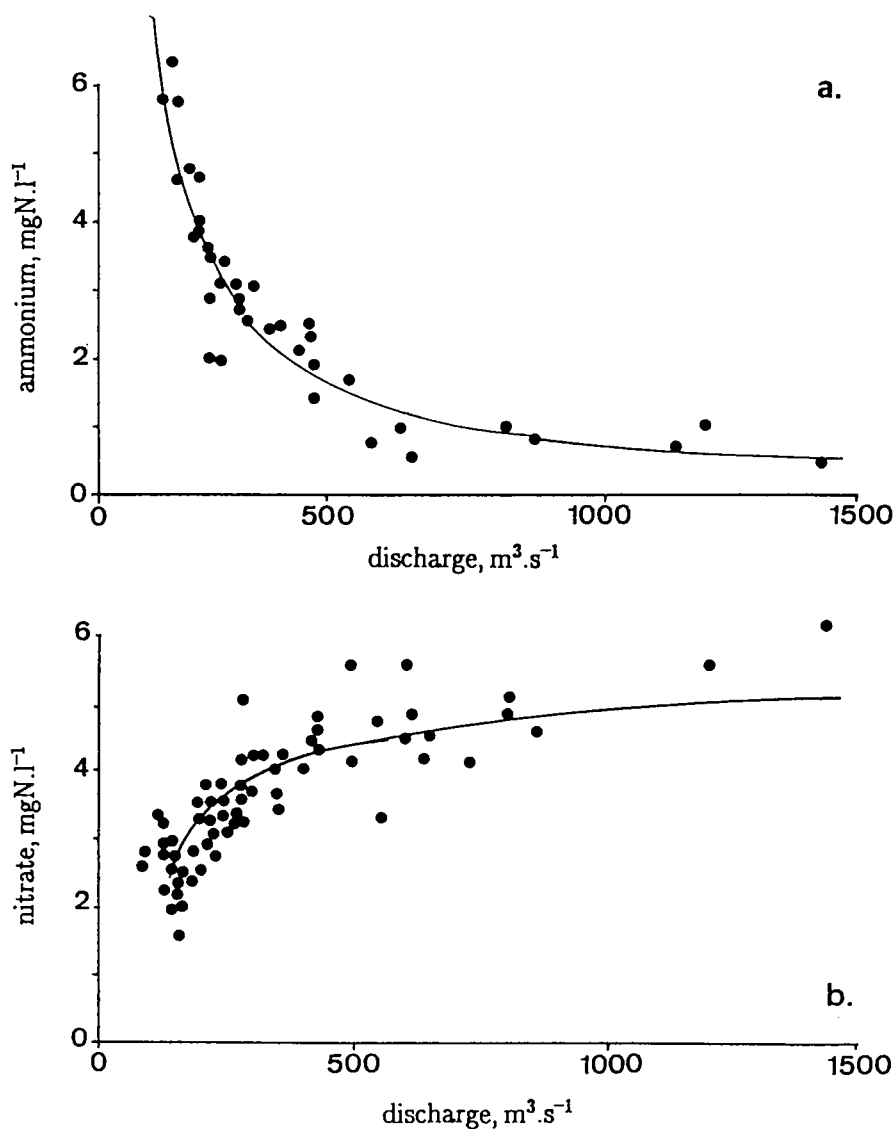


Fig. 2. NH_4 (a) and NO_3 (b) concentrations in the lower Seine River, plotted against river flow [data from AFSN (1983–1987) and our own measurements (1989–1990)].

This relation indicates the overwhelming predominance of point sources in the origin of ammonium. From these data, a mean daily load of ammonium discharged into the Seine River above Méricourt can be evaluated as about 70 tons N.

NO₃ concentration on the other hand follows an increasing hyperbolic relation with discharge (Fig. 2b), showing that non point sources dominate. Indeed, wastewater from Achères is devoided of nitrate and does not affect NO₃ concentration in the river, except by slight dilution. The relationship observed between nitrate and discharge can be interpreted as the result of the mixing of two types of upstream water, each with its own nitrate concentration (Johnson et al. 1969). Thus, the instantaneous discharge, Q, of the Seine river can be considered as the sum of a nearly constant baseflow component, Q_b and a more variable surface runoff component Q_s. If constant nitrate concentrations, N_b and N_s respectively, are associated with these two components of the river discharge, the resulting nitrate concentration of the river water is expected to obey the relationship:

$$\text{NO}_3 = \text{Nb} (\text{Qb}/\text{Q}) + \text{Ns} (\text{Qs}/\text{Q})$$

or

$$\text{NO}_3 = \text{Ns} - (\text{Qb}/\text{Q}) \cdot (\text{Ns} - \text{Nb})$$

Best fitting of this relationship with the experimental data provides the following values of the parameters:

$$\text{Qb} = 150 \text{ m}^3/\text{s}$$

$$\text{Nb} = 2.8 \text{ mgN/l}$$

$$\text{Ns} = 5.1 \text{ mgN/l}$$

This interpretation is supported by the observation of the short term variations of nitrate concentration during a flood in Paris from 10 to 24 February 1990 (Fig. 3): the rapid rise of discharge above a level close to base flow was accompanied by a parallel increase in nitrate concentration, up to 6 mgN/l.

Longitudinal profiles of nitrate and ammonium

At several occasions under conditions of low and stable flow, data were obtained on the variations of nitrate, ammonium and organic nitrogen concentration within a single water mass carefully followed as explained in

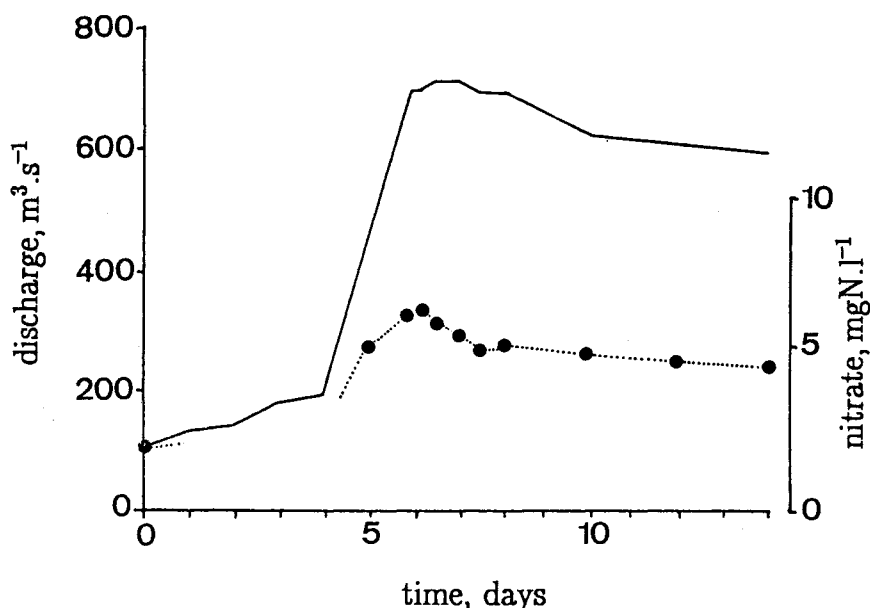


Fig. 3. Short term variations of nitrate concentration (dotted line) during a flood of the Seine in Paris (February 10–24, 1990). Solid line represents discharge.

Methods during its downriver travel in the Seine below the outfall of the Achères plant and the confluence of the Oise river. Typical results are presented in Fig. 4. Data from instantaneous samplings in the upstream portion of the river are also shown.

Total inorganic nitrogen remains generally very stable or increases slightly over the entire 100 km portion of the river downstream Achères. Nitrate concentration remains essentially constant down to Méricourt, about 50 km from Achères. It is not significantly affected by the discharge of the treatment plant, nor by the confluence of the Oise River. It then increases by about 1 mgN/l between Méricourt and Poses. Ammonium concentration increases considerably due to the discharge of Achères. When the flow from the Oise River is significant with respect to the upstream flow from the Seine River, a dilution is observed at the confluence. It then remains constant down to Méricourt and slightly decreases in the lowest stretch of the river. Organic nitrogen represents a small part of total nitrogen load. It only decreases slowly below Achères. Part of this decrease is probably due to sedimentation of particulate organic matter.

The data available from the samplings carried out by the AFBSN, although they do not represent rigorous follow up of a same water mass, presents the same general trends (see below, Fig. 9). In particular the

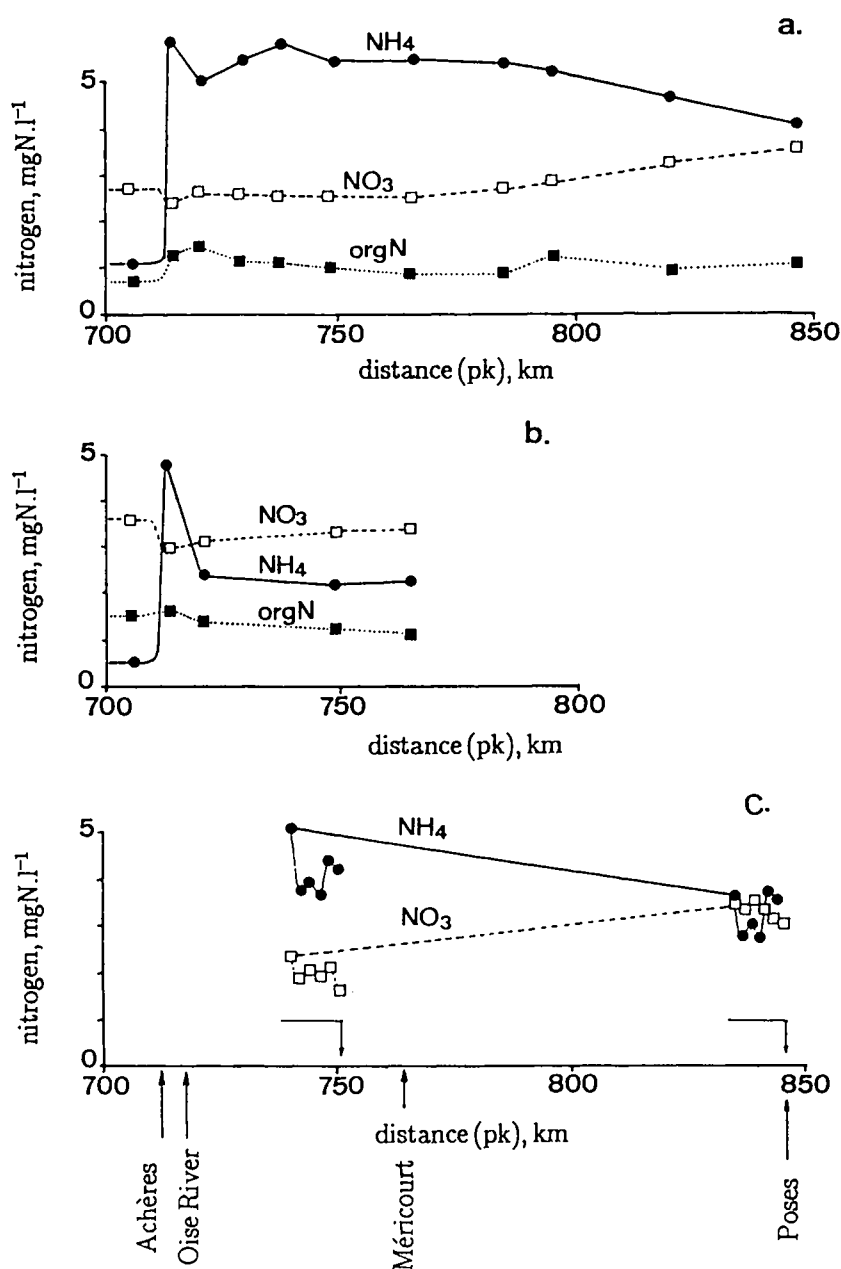


Fig. 4. Longitudinal profiles of nitrogen species obtained by carefully following a water mass during its downriver travel from below the Achères sewage treatment plant: (a) October 1989, (b) June 1989, (c) June 1990. The data from the latter situation includes the observation over 24 h of the corresponding variations of nitrate and ammonium at two fixed stations (Porcheville and Poses).

slight increase of nitrate and decrease of ammonium concentration below Méricourt is systematically observed. The rate of ammonium reduction between Méricourt and Poses calculated from these data has been plotted against residence time of the water masses in the same river stretch for all available profiles corresponding to situations with water temperature above 15 °C (Fig. 5). The results indicate a rate of ammonium consumption in the range 0.0075–0.02 mgN.l⁻¹.h⁻¹.

Abundance and activity of nitrifying bacteria

Longitudinal profiles of MPN numbers and activity of nitrifying bacteria are shown in Fig. 6. Upstream Paris and down to Andrésy, both abundance and activity are very low. A significant increase occurs in the lowest part of the river, where nitrification rates between 0.007–0.015 mgN.l⁻¹.h⁻¹ are recorded. This range is exactly that required for explaining the observed decrease of ammonium and increase of nitrate in the reach Méricourt-Poses (see Fig. 5), showing that nitrification in the water column explains most of the transformations of inorganic nitrogen in the river. The delay in the development of nitrifying bacteria following the increase in ammonium concentration at of Achères is consistent with the

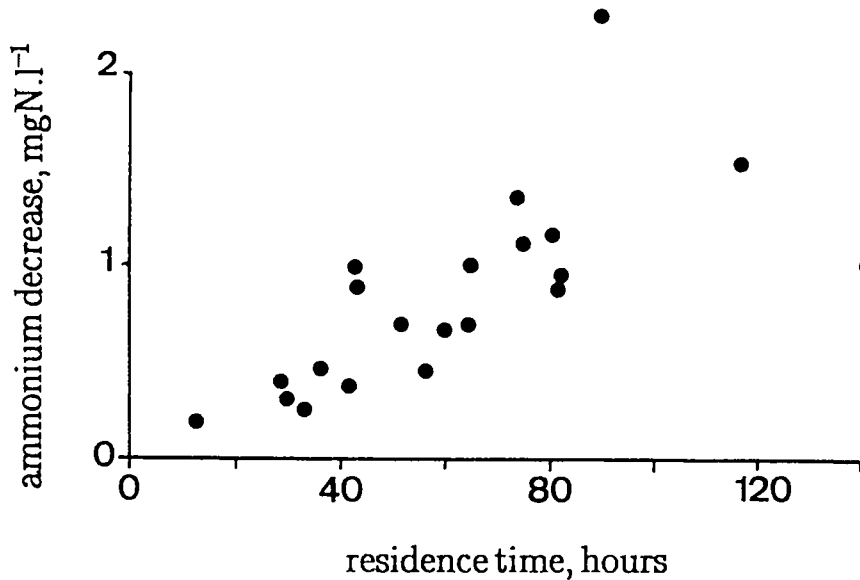


Fig. 5. Decrease in ammonium concentration observed between Méricourt (pK 765) and Poses (pK 846), as a function of residence time (estimated from river discharge) during summer months (temperature ≥ 15 °C) (data AFBSN).

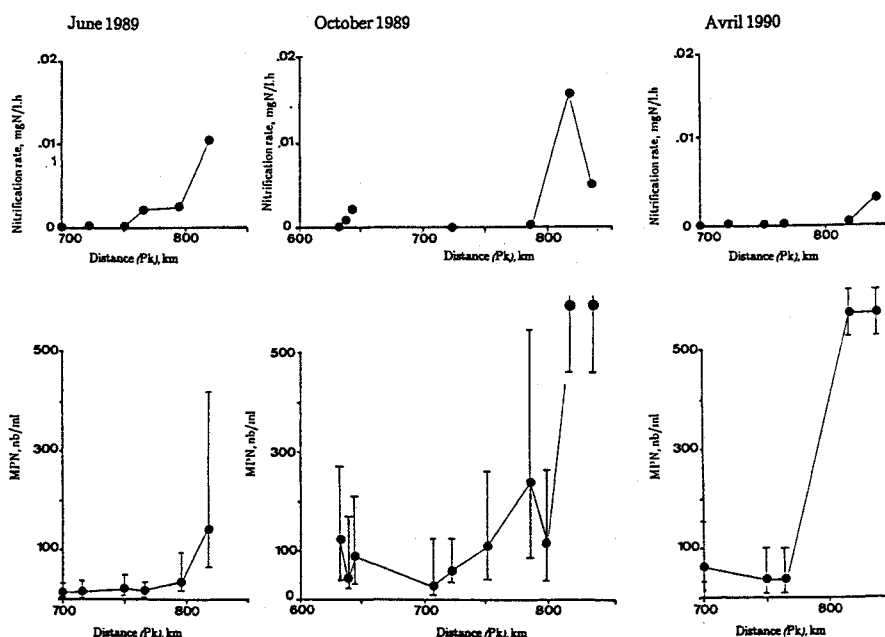


Fig. 6. Longitudinal profiles of abundance (lower panel) and activity (upper panel) of nitrifying bacteria in the river Seine in June 1989, October 1989 and June 1990.

observation that no increase in nitrate nor decrease in ammonium are observed in the first 50 km below the outfall of the water treatment plant. Roughly, the increasing trend in numbers of nitrifying bacteria observed in the 3 profiles between Achères and Poses, corresponds to an apparent growth rate of about 0.02 h^{-1} .

Benthic fluxes

A few *in situ* measurements of the fluxes of nitrate and ammonium across the sediment-water interface were carried out between Achères and Poses during periods of low flow. A typical example of the data obtained is presented in Fig. 7. Systematically, nitrate consumption within the bell-jar was observed, while ammonium concentration increased. Nitrate consumption was likely the result of benthic denitrification in these anoxic sediments. The production of ammonium resulted from the ammonification of sedimented organic material. After 2–4 hours incubation in summer, 10 to 20 hours in winter and spring, oxygen was exhausted from the bell-jar. Often, this resulted in a decrease in the rate of NH_4 production and an increase in NO_3 reduction. Table 1 gathers the flux rates

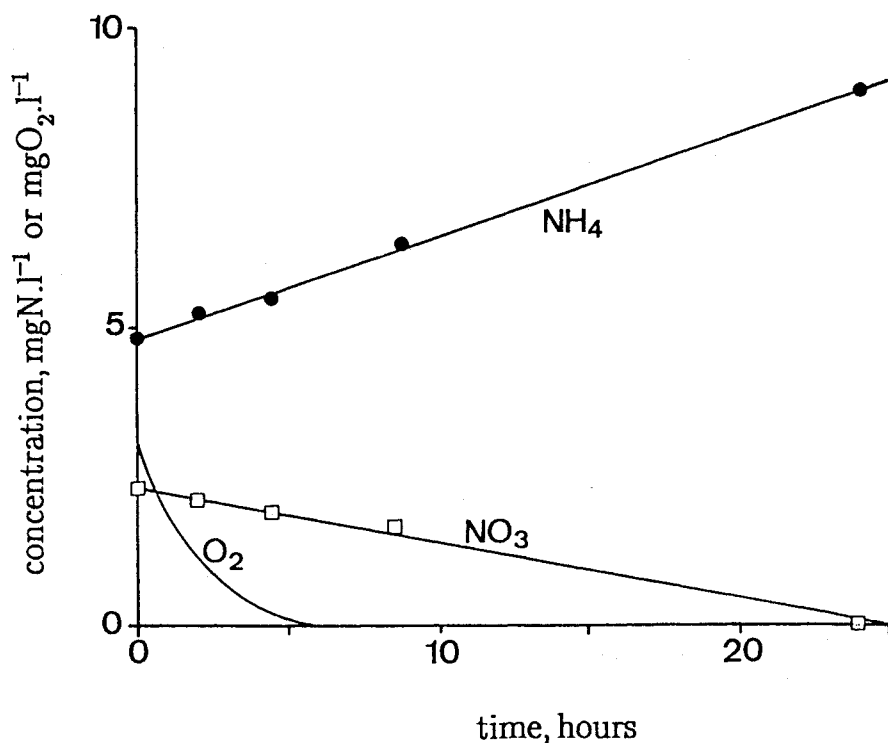


Fig. 7. Typical result of bell-jar experiments intended to evaluate the fluxes of nitrate and ammonium across the sediment-water interface: long term variations of nitrate, ammonium and oxygen concentration within the bell-jar, after it has been deposited on the bottom (Porcheville, September 1990).

estimated from the data of the 1–4 first hours of bell-jar incubation, available for 3 stations of the river. Measurements obtained close to the outlet of the purification plant of Achères show the highest values of the fluxes. The fluxes of nitrate consumption are quite comparable to those measured in the sediments of a storage basin fed with the water of the Oise River (Billen et al. 1989). The fluxes of ammonium are at least ten times higher, however, indicating the higher organic content of the Seine sediments. Considering that about one third of the channel bottom area between Achères and Poses is covered by muddy sediments similar to those on which the bell-jar experiments were carried out (as estimated on the basis of observations made on a few cross section of the river), total net benthic production of ammonium and consumption of nitrate in this sector during summer conditions can be evaluated to about 9 TN-NH₄.d⁻¹ and 3 TN-NO₃.d⁻¹ respectively.

Table 1. Measurement of NH_4 and NO_3 flux across the sediment water interface measured with a belljar deposited on the bottom of the river Seine at 3 stations (negative values indicate flux directed to the sediments).

	temperature °C	NH_4 flux $\text{mgN/m}^2\cdot\text{h}$	NO_3 flux $\text{mgN/m}^2\cdot\text{h}$
Achères (pK 708)			
March 5, 1991	10	143	-39.4
May 21, 1991	18.5	660	-14.5
Porcheville (pK 750)			
June 14, 1990	18	86	-5.9
Sept 4, 1990	21.5	33	-8.2
March 6, 1991	9	22	-10.7
May 23, 1991	18	53	-20.7
Poses (pK 846)			
June 21, 1990	19	69	-38.5

Discussion

The data presented in this paper allows to establish a tentative budget of nitrogen transport and transformation in the river Seine below Paris under conditions of low discharge characteristic of the summer period (Fig. 8). Fluxes of the various nitrogen species from the upstream course of the Seine river, the treatment plant of Achères and the Oise river were estimated by multiplying the corresponding flow rate values with the measured concentrations. Water column ammonification and phytoplankton uptake were calculated from estimates of bacterial activity and net primary production data carried out by Servais et al. (1990) and Garnier et al. (1990) respectively, considering a mean C/N ratio of 7 for organic matter.

In spite of the uncertainty remaining on the quantitative determination of several of the processes involved, Fig. 8 offers a clear and coherent picture of the nitrogen cycling in the river Seine. The rather slow reactivity of most nitrogen forms in this stretch of the river is striking. Nitrification is the most important internal process affecting nitrogen within this lower part of the river. It only starts about 20 km before the estuarine zone, where most of the ammonium load remains to be oxidized. Nitrification is indeed a major factor in the oxygen balance of the Seine estuary (Romana, pers. comm.), while it plays only a minor role as a factor of oxygen depletion in the river itself. The reason for the lack of an intense nitrifica-

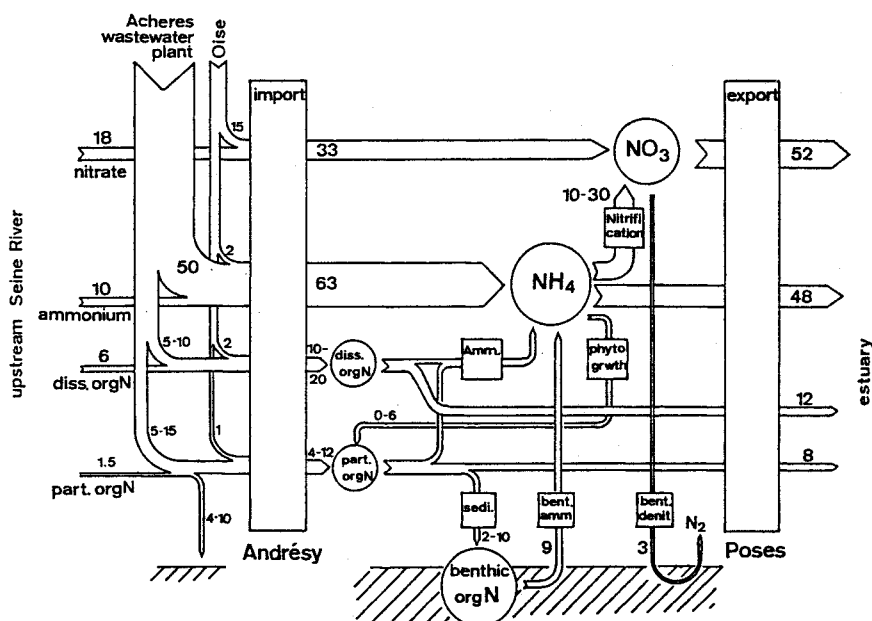


Fig. 8. Tentative budget of nitrogen transport and transformation in the lower Seine River under representative summer, low flow conditions ($180 \text{ m}^3/\text{s}$). Figures are given in 10^3 kgN per day.

tion in the river is likely to be explained by the slowness of the nitrifiers growth.

The upstream course of the river carries very few nitrifying bacteria. The kind of treatment presently applied in the wastewater plant of Achères does not result in the discharge of a significant population of nitrifying bacteria (contrarily to what is observed for heterotrophic bacteria [Garnier et al. 1992]). Therefore, only very low numbers of nitrifying bacteria are present in the river. Even though ammonium is present below the outfall of Achères at concentrations far above the requirement for optimal growth of nitrifying bacteria, they take several days to reach a population level able to carry out a significant activity of ammonium oxidation. Indeed, the apparent growth rate of nitrifying bacteria observed between Achères and Poses (about 0.02 h^{-1} at temperatures ranging from 14 to 20°C) is very close to the reported maximum growth rate for *Nitrosomonas* (0.036 at 25°C , with a Q_{10} of 3.3, according to Helder & De Vries (1983), who considered these values as rather general for brackish and freshwater environments).

Denitrification also appears of little significance in the nitrogen budget of this part of the Seine river. It only occurs in the benthic phase at rates

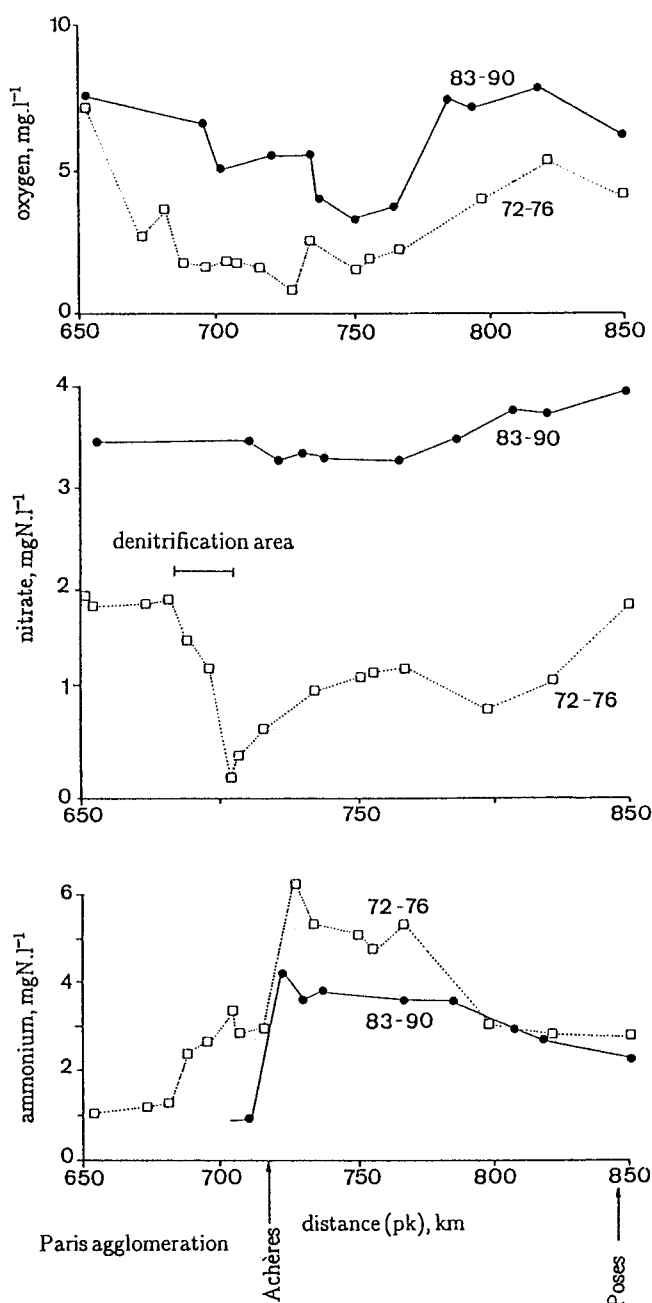


Fig. 9. Mean longitudinal profile of nitrate concentration as observed during the period 1972–1976 under low flow and high temperature conditions (after Lesouëf & André, 1982). Comparison with the mean present day summer situation (period 1983–1990).

which are not high enough to greatly affect nitrate transport by the river. The restoration of good oxygen conditions in the lower Seine, owing to the extension of the sewage treatment plant of Achères at the end of the 70's led indeed to the disappearance of an area of intense denitrification in the water column itself. Previously, this process was responsible for the elimination of a significant part of the nitrate load from the upstream course of the Seine. This is nicely shown in the data published by Lesouëf & André (1982) (Fig. 9). These authors calculated the mean oxygen, nitrate and ammonium concentrations at several stations on the Seine River for all low flow (discharge at Paris lower than $100 \text{ m}^3 \cdot \text{s}^{-1}$) and high temperature (above 15°C) situations during the period 1973–1976, i.e. before the extension of the Achères treatment plant. Using the same criteria, we calculated the corresponding mean longitudinal profiles from our summer data for the period 1983–1990 (Fig. 9). The comparison confirms the present absence of a significant denitrification in the Seine River below Paris. It also shows that the nitrate load presently carried down by the river Seine has increased, while ammonium concentration has remained approximately constant. This increase is partly the result of higher upstream NO_3 concentration, associated with increased fertilisation and leaching of agricultural soils. A part of the increase, however, is the result of the fact that the process of denitrification in the Seine below Paris is now much more limited owing to better oxygen conditions in the river. This indicates that the extension of standard activated sludge wastewater treatment without tertiary nutrient elimination, although it resulted in restoring better oxygen conditions in rivers, paradoxically resulted in an increase, instead of a reduction, of the nitrate load carried down to the sea.

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