Dissolved Loads of Rivers and Surface Water Quantity/Quality Relationships
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Dissolved and suspended loads of the regulated River Nidda in the Rhine-Main area

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ABSTRACT Over a period of two years, the daily load of macro- and trace constituents of the regulated River Nidda and its tributaries were analysed for the liquid and solid phases and the bed sediment. Parameters analysed were runoff, total suspended solids (TSS), suspended sediment-bound heavy metals, total dissolved solids (TDS), dissolved constituents such as Ca^{2+}, Mg^{2+}, Na^+, K^+, Cl^-, HCO_3^-, SO_4^{3-} and the heavy metals Cd, Cu, Ni, Co, Cr, Pb, Zn. The TSS/TDS relationship displays the dominance of dissolved loads for low water periods and winter floods. For summer floods, primarily generated by heavy cloudbursts, the TSS/TDS relation demonstrates the TSS to be the main load during rising stage. The maximum TSS load does not coincide with peak discharge. The heavy metal loads in the solute and solid phases are low compared with polluted rivers of the FRG, and the same situation exists for the concentrations of heavy metals in bed sediment. An important source for heavy metals are storm water flows from tributaries which drain suburban or urban areas.

Substances dissolutes et en suspension dans le bief inférieur régularisé de la Nidda

RESUME Durant une période de deux ans la charge journalière de matières en suspension et dissoutes de la Nidda régularisée et de ses tributaires a été analysées dans la phase liquide, la phase solide et les sédiments sur le fond du lit. Les paramètres analysés sont les suivants: l'écoulement, la charge totale solide en suspension (TTS), les métaux lourds en état d'adsorption absorbés par les sédiments, la charge dissoute totale (TDS), les substances dissoutes telles que Ca^{2+}, Mg^{2+}, Na^+, K^+, Cl^-, HCO_3^{2-}, SO_4^{2-}, PO_4^{3-} et les métaux lourds Cd, Cu, Ni, Co, Cr, Pb, Zn. La relation TSS/TDS montre la prédominance des charges dissoutes pendant la période de basses eaux et pendant les crues hivernales. Quant aux crues estivales, principalement dues aux pluies torrentielles, la relation TSS/TDS montre que le TSS est le constituant principal au moment de la montée de l'eau. Le chargement maximum en substances TSS ne coincide pas avec la pointe de crue. La quantité des métaux lourds dans la phase liquide aussi bien que dans la phase solide est faible comparativement à celle des rivières polluées de la RFA, de même pour la concentration des métaux lourds dans la sédimentation de fond. Une source importante pour les métaux lourds se trouve dans les eaux de pluies apportées par le système des égouts suburbains et urbains.

THE RIVER NIDDA DRAINAGE BASIN

The River Nidda, a tributary of the Main, drains a basin of about 1950 km^2, about 870 km^2 of which are uplands (400-800 m a.s.l.) of the Vogelsberg (600 km^2) and the Taunus (270 km^2). The main land use of the uplands are range, pasture (240 km^2) and forest (497 km^2). The dominant forest type is a deciduous forest (223 km^2). The rest of the area is covered with coniferous forest (164 km^2) and mixed forest (109 km^2). The principle land use of the Wetterau (1080 km^2), a loess covered basin (100-300 m a.s.l.), is agriculture (979 km^2). About 234 km^2 of the total area of the basin is urban or suburban land and a portion of this represents impervious areas (Fig.1). The headwaters of the River Nidda and its tributaries drain the uplands of the Vogelsberg and the Taunus with steep longitudinal gradients, while slopes of the river channels are quite gentle in the Wetterau and the lower reaches. The headwaters of the River Nidda system flow in natural beds, but the middle and the lower reaches are canalized and from the mouth to the gauge at Bad Vilbel are regulated by six weirs. Seven rivulets draining the Taunus ranges are tributary to the regulated river section as are a number of urban and suburban storm water inlets.

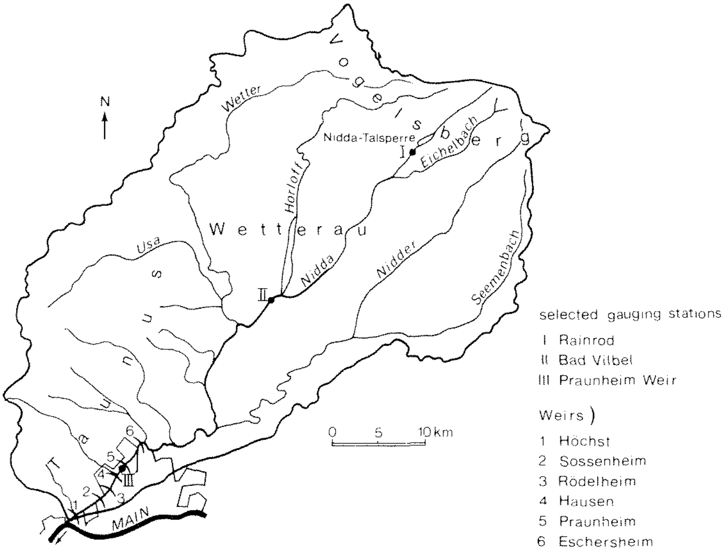

FIG.1 The River Nidda drainage basin.

In general, runoff conditions are such that winter floods, dependant on rainfall and snowmelt, occur from November to March, whereas serious flash floods are frequent during summer months and are generated by heavy storms and cloud bursts. Low water conditions are typical in summer months and autumn. Under serious drought conditions (e.g. in 1976), numerous wells and sections of the headwaters become dry. The near-surface groundwater aquifers are widely exhausted for parts of the basin so that interflow and baseflow are negligible contributors to runoff. Under such circumstances, the primary sources of runoff are the baseflow from the basalt aquifers of the Vogelsberg, the River Nidda reservoir at Rainrod and the waste return flow.

MATERIAL AND METHODS

Over a period of 2 years the daily loads of macro- and trace constituents of the lower River Nidda and its tributaries were analysed for the liquid and solid phases and the bed sediment. Parameters analysed in this particular study were runoff (Q), total suspended solids (TSS), suspended sediment-bound heavy metals, total dissolved solids (TDS), dissolved constituents such as Ca^{2+}, Mg^{2+}, Na^+, K^+, Cl^-, HCO_3^-, SO_4^{2-}, PO_4^{3-}, heavy metals and selected bed sediment-bound constituents. Sampling procedures, conservation measures and transport of samples (water, suspended sediment, bed sediment) were in accordance with the methods proposed by the US National Handbook (US Dept of the Interior, 1977). Analyses were done by titration (Cl^-, HCO_3^-), photometry (SO_4^{2-}, PO_4^{3-}) and atomic absorption spectrometry (AAS) for the heavy metals. Samples were subjected to acid digestion (HNO_3 conc.) before AAS determination.

WINTER FLOODS

Flood conditions in the regulated River Nidda during the cold season from November to March are characterized by a consecutive number of flood waves generated by rainfall, rain-on-snow, snowmelt or more frequently by combinations of these flood generating mechanisms.

A flood in January 1982 (29 December 1981-18 January 1982, Table 1) displays the typical hydrograph of winter floods. A number of floods occurred in the River Nidda during November and December 1981 but low water conditions, following the falling limb of a flood event on 8-10 December 1981, had been established immediately preceding the

TABLE 1 Hydrograph of a winter flood during December 1981 and January 1982 in the regulated River Nidda at Praunheim weir

Date	*Runoff ($m^3 s^{-1}$)*	*Date*	*Runoff ($m^3 s^{-1}$)*	*Date*	*Runoff ($m^3 s^{-1}$)*
29	12.4	5	113.0	12	28.4
30	13.8	6	101.0	13	22.9
31	37.5	7	88.0	14	13.7
1	69.7	8	75.3	15	16.5
2	63.9	9	63.9	16	15.1
3	65.8	10	50.4	17	13.7
4	119.0	11	30.1	18	12.4

January 1982 event (30 December 1981). The latter flood resulted from a combination of rain, rain-on-snow and snowmelt, but these different sources predominated during different sections of the flood hydrograph. The pre-flood low water and the low flows at the end of the falling limb of the flood hydrograph display typical values of suspended and dissolved loads, characteristic differences in the concentrations of dissolved constituents and a TSS/TDS ratio of 0.03, which indicates the source of runoff to be mainly baseflow, interflow and some waste water. The pH level was 7.2 in these conditions. The flood peak was generated by rainfall totals of *c.* 20-25 mm, and some snowmelt from the middle reaches of the drainage area. The TSS/TDS ratio was 0.5-0.6 and reflects the entrainment of a great amount of bed sediment into the flood wave. The detachment of material by soil erosion processes in the basin did not significantly contribute to the suspended load. The pH level was *c.* 6.1-6.3, which is considered to be a normal level of hydrogen ion concentration in winter floods.

TABLE 2 Total runoff ($m^3 5min^{-1}$), total suspended solids (kg $5min^{-1}$) and total dissolved solids (kg $5min^{-1}$) expressed by selected dissolved constituents analysed at Praunheim weir during pre-flood conditions, rising wave, peak discharge and falling wave in January 1982

Date	*Runoff*	Ca^{2+}	Mg^{2+}	Na^+	K^+	Cl^-	HCO_3^-	SO_4^{3-}	PO_4^{3-}	*TSS*	*TSS/ TDS*
30	*4 140*	*337*	*84*	*294*	*45*	*546*	*944*	*91*	*5.5*	*79*	*0.03*
31	*9 450*	*933*	*236*	*804*	*118*	*1485*	*2576*	*225*	*14*	*248*	*0.04*
4/5	*35 700*	*1815*	*465*	*810*	*150*	*1635*	*2190*	*390*	*30*	*3510*	*0.47*
6	*30 200*	*1530*	*390*	*660*	*150*	*1260*	*1500*	*270*	*30*	*3480*	*0.60*
7	*26 400*	*1275*	*300*	*540*	*90*	*1065*	*12*	*240*	*24*	*2835*	*0.80*
11	*9 030*	*600*	*150*	*270*	*60*	*495*	*285*	*150*	*8*	*990*	*0.49*
20	*3 720*	*307*	*72*	*238*	*24*	*438*	*810*	*83*	*6*	*66*	*0.03*

No rainfall occurred in the period from 6 to 8 January and the principal source feeding runoff was snowmelt. A very sensitive parameter for detecting the contribution of melt to flood flow is the hydrogen ion concentration. Meltwater is strongly acid in areas where wet and dry deposition of atmospheric SO_x and NO_x compounds are high, which is the case in the River Nidda drainage basin (Perseke, 1982). In consequence, the pH level of the river water decreased dramatically in this period to a value of 3.0-3.5. However, since quickflow from snowmelt eroded some soil material, the TSS/TDS ratio also increased to 0.8. Total loads of suspended and dissolved solids for the pre-flood situation as well as for the entire flood hydrograph at Praunheim weir are presented in Table 2.

It is generally agreed that the suspended load carried by rivers is mainly controlled by turbulent mixing dynamics between the water and sediment phases, if detachment and erosion of material from the drainage basin is negligible (Bogardi, 1956; Ward, 1978). For winter floods, the suspended and dissolved loads of the River Nidda at

Praunheim weir increase in relation to discharge. The relationship of total runoff (Q) ($m^3 5min^{-1}$) and TSS (kg $5min^{-1}$) for the January flood at rising stage is:

$$TSS = 2.48 \times 10^{-5} Q^{1.78} \qquad r^2 = 0.99 \tag{1}$$

and the same relationship for falling stage is:

$$TSS = -160.16 + 0.11 Q \qquad r^2 = 0.98 \tag{2}$$

In the case of dissolved solids loads, the runoff/TDS relationship as well as the relation between runoff and different constituents and the distribution pattern during flood conditions are similar to those of the runoff/TSS relationship, but total loads differ considerably in accordance with the sources from which they are derived. If constituents are undergoing chemical reactions (e.g. the dependence of HCO_3^- load on pH conditions) or the compounds are mainly supplied from waste water inflows or road de-icing procedures, no relation between runoff and the dissolved elements is found. As a rule, a coincidence of peak discharge and maximum loads of TSS and TDS was observed for winter floods.

SUMMER FLOODS

Heavy thunderstorms are responsible for the sometimes disastrous floods during summer months in the middle and lower reaches of the River Nidda system. In summer floods, the suspended and dissolved loads of the River Nidda are controlled by turbulent mixing of water and sediment during the flood wave and the erosional power of the quickflow in the drainage area, which is closely related to vegetation cover, land use and relief; and by the main sources for dissolved substances, which are surface leaching and a retarded interflow-baseflow contribution.

A flood hydrograph in August 1981 (8-26 August 1981, Table 3) displays the characteristic features of a summer flash flood in the River Nidda system. A tremendous thunderstorm in the middle and lower reaches of the River Nidda provided rainfall totals of *c.* 100-150 mm in a couple of hours. A great amount of rainwater entered

TABLE 3 Hydrograph of summer flood in August 1981 in the regulated River Nidda at Praunheim weir

Date	*Runoff* *($m^3 s^{-1}$)*	*Date*	*Runoff* *($m^3 s^{-1}$)*	*Date*	*Runoff* *($m^3 s^{-1}$)*
8	*7.8*	*14*	*80.2*	*20*	*40.1*
9	*12.8*	*15*	*70.5*	*21*	*32.6*
10	*73.6*	*16*	*72.2*	*22*	*28.9*
11	*100.0*	*17*	*46.1*	*23*	*25.4*
12	*95.0*	*18*	*38.2*	*24*	*23.3*
13	*83.0*	*19*	*40.1*	*25*	*21.1*

the direct flow and built up a serious flash flood which spilled over the banks. The immediate pre-flood situation was characterized by lowflow conditions with a TSS/TDS relation of 0.0004, which demonstrates the TSS to be at the very minimum. The same conditions were observed at low water stage at the end of the falling limb of the flood hydrograph 15 days after peak discharge. The water was slightly alkaline with the hydrogen ion concentration ranging from pH 8.5 to 8.7. Some hours after the initial rainfall and during rapidly rising stage, the direct flow carried a great amount of eroded material from the basin area and turbulence promoted the uptake of even coarse bed sediment into the flood wave. The TSS/TDS relationship changed drastically to 3.5, while the hydrogen ion concentration decreased only slightly to pH 7.5, which demonstrates the buffering power of deposited bed sediment and the soils of the drainage basin. A flushing out by quickflow was evident for Mg^{2+} and PO_4^{3-} ions. Other dissolved solids were in part leached from the basin in surface runoff, but a significant amount of solutes were released from the zones drained by interflow and baseflow after soil pores and the near-surface groundwater aquifers had been filled up. The flow time characteristics of the drainage area and the retardation of runoff in interflow and baseflow were responsible for the lag in the transport of dissolved solids from the basin. The maximum loads of several constituents occurred one or two days after peak runoff. The TSS/TDS ratio at this time was *c.* 0.5 and the pH values were constant at 7.5-7.7. Six days after peak runoff, the TSS/TDS ratio decreased to 0.09, while the hydrogrn ion concentration increased to pH 8.4. Both parameters reflect the increasing contribution of solutes in interflow and baseflow and the dominance of dissolved solids during falling stages.

A representative set of data for runoff, TSS and TDS before, during and after peak runoff for a serious flood in August 1981 is presented in Table 4. Except for Mg^{2+} and PO_4^{3-} loads, the dissolved solids did not coincide with TSS during rising stage, when maximum TSS load occurred before the runoff peak. The relationship of total runoff ($m^3 5min^{-1}$) and TSS ($kg\ 5min^{-1}$) for the August flood during rising stage is:

$$TSS = 2.19 \times 10^{-22}\ Q^{5.94} \qquad r^2 = 1.0 \qquad (3)$$

and the same relationship for falling is:

$$TSS = 7.28\ Q^{2.37\times 10^{-4}} \qquad r^2 = 0.79 \qquad (4)$$

LOW WATER CONDITIONS

In low water periods, the principal sources of runoff are baseflow and waste water discharges, which return water used for industrial or domestic purposes to the river either directly or after passing through a treatment plant. Turbulent mixing of sediment and water at low stages is negligible, the most important sources of suspended solids are waste water discharges and, especially in the regulated section of the River Nidda, the production of phytoplankton. The latter source becomes more important if atmospheric controls of

TABLE 4 Total runoff ($m^3 5min^{-1}$), total suspended solids ($kg\ 5min^{-1}$) and total dissolved solids ($kg\ 5min^{-1}$) expressed by selected dissolved constituents analysed at Praunheim weir during pre-flood conditions, rising wave, peak discharge and falling wave in August 1981

Date	Runoff	Ca^{2+}	Mg^{2+}	Na^+	K^+	Cl^-	HCO_3^{2-}	SO_4^{2-}	PO_4^{3-}	TSS	TSS/TDS
9	3 840	90	24	120	19.5	25.5	390	150	13.5	0.3	0.0004
10	22 080	540	114	390	186	810	840	840	90	13 200	3.46
11	30 000	915	225	375	210	990	1320	1395	75	2 730	0.50
12	28 500	1020	300	330	150	810	615	1020	75	2 160	0.50
16	21 600	810	210	240	90	570	1620	930	30	390	0.09
26	6 180	390	120	180	30	420	750	240	12	1.5	0.0005

TABLE 5 Average runoff (m^3s^{-1}) and average suspended and solute loads ($kg\ s^{-1}$), and total runoff ($m^3\ 5\ min^{-1}$) and total suspended and solute loads ($kg\ 5\ min^{-1}$) at Praunheim weir for a low water period of 18 days in September 1981

	Runoff	Ca^{2+}	Mg^{2+}	Na^+	K^+	Cl^-	HCO_3^-	SO_4^{2-}	PO_4^{3-}	TSS	TSS/TDS
Average	8.4	0.69	0.20	0.56	0.08	1.01	1.44	0.54	0.02	0.02	-
Standard deviation	0.56	0.08	0.00	0.09	0.01	0.07	0.42	0.14	0.004	0.02	-
Total	2520	270	60	168	24	303	432	162	6	6	0.004

production, mainly solar radiation, are favourable (Bernerth & Tobias, 1982). Another important parameter ruling the efficiency of primary production is residence time, which is sufficiently high at low water stages.

A representative collection of data on suspended and dissolved loads for a low water period of 18 consecutive days in September 1981 is presented in Table 5. The TSS/TDS ratio is 0.004, which demonstrates the dominance of dissolved loads at low water stage. The suspended solids are primarily in the organic phase (e.g. waste waterborne or phytoplankton). The hydrogen ion concentration level was pH 8.8 and may increase to 10.5 at maximum on days when river water is fully exposed to solar radiation, which strongly suggests the influence of biogenic activity in the shallow lake-like regulated river section. There was no relationship found between total runoff (m^3 $5min^{-1}$) and TSS (kg $5min^{-1}$), which is expected because the primary sources of TSS at low stages are runoff independent variables such as waste inputs and primary production.

TRACE POLLUTANTS

The source of a considerable amount of the heavy metals in the liquid phase of the regulated River Nidda is geochemical (e.g. metals are leached out of soils and bedrock). Mainly for Zn and Pb, and to a lesser extent for Cu, Cr and Cd background concentrations from geochemical sources are supplemented by waste water discharges, by flushing out of polluted areas with high traffic densities (mainly roads and parking lots) and by the leaching of cultivated soils, which are heavily treated with mineral fertilizers. Some data on the quantity of heavy metals in solution are given in Table 6.

TABLE 6 Heavy metal loads (g $5min^{-1}$) in the solute phase at Praunheim and Rödelheim weirs under different flow conditions

Heavy metals	*Floodwater runoff*	*Average flow*	*Minimum flow*
Cd	*3*	*0.5*	*0.09*
Cu	*180*	*28*	*5.4*
Ni	*240*	*38*	*7.2*
Co	*30*	*4.8*	*0.9*
Cr	*60*	*9*	*2.2*
Pb	*90*	*14*	*2.7*
Zn	*1260*	*201*	*36*

The bed sediments in the lower River Nidda are in an unstable state because flow regulation by the six weirs is responsible for complex uptake, transport and deposition mechanisms. With the weirs open, the flood waves of winter and summer entrain and export the bulk of bed sediment deposited. Since residence time for bed sediments is low, the enrichment of heavy metals is practically non-existent in this section of the river. The concentrations of heavy metals in different types of sediment are summarized in Table 7.

TABLE 7 Average concentration (mg kg^{-1}) of heavy metals in bed sediment near to a storm water intake (A), bed sediment (B), flood deposited sediment (C), in the regulated River Nidda

Heavy metals	*A*	*B*	*C*
Cd	*1.8*	*1.8*	*0.8*
Cu	*61*	*65*	*46*
Ni	*37*	*37*	*40*
Co	*12*	*9*	*11*
Cr	*50*	*45*	*42*
Pb	*102*	*88*	*58*
Zn	*510*	*346*	*268*

Suspended sediments are mainly derived from bed sediment, which is entrained in the flood wave by turbulent mixing of water and sediment; from the drainage area, where eroded materials are channelled into the river by direct flow, and from waste water discharges and primary production, which are sources of importance under low water conditions. Depending on which of these sources is dominant the suspended matter contains heavy metals which differ in concentration and composition. Selected data of loads of suspended sediment-bound heavy metals are given in Table 8.

Urban and suburban areas are largely impervious. The material deposited in these areas during dry periods is flushed out by rain. A number of suburban storm water drains flow directly into the regulated River Nidda and produce a considerable amount of TDS, TSS and suspended sediment-bound heavy metals. The main data for storm water discharges are presented in Table 9. For total storm water

TABLE 8 Heavy metal loads (g $5min^{-1}$) in the suspended phase at Praunheim and Rödelheim weirs under different flow conditions

Heavy metals	*Floodwater runoff*	*Average flow*	*Minimum flow*
Cd	*2.2*	*0.06*	*0.009*
Cu	*213*	*5.5*	*0.09*
Ni	*66*	*17*	*0.03*
Co	*11*	*0.3*	*0.004*
Cr	*33*	*0.9*	*0.01*
Pb	*121*	*3.2*	*0.05*
Zn	*960*	*25*	*0.4*

TABLE 9 Load data for suspended sediment (kg $5min^{-1}$) and suspended sediment-bound heavy metals (g $5min^{-1}$) in a suburban storm water system draining into the regulated River Nidda

	Maximum load	*Average maximum load*	*Average minimum load*	*Minimum load*
Sediment	*120.1*	*34.5*	*3.3*	*0.4*
Heavy metals:				
Cd	*2.6*	*0.68*	*0.07*	*0.001*
Cu	*58.5*	*13.2*	*1.6*	*0.007*
Ni	*12.8*	*3.3*	*0.3*	*0.001*
Co	*3.9*	*0.7*	*0.05*	*0.001*
Cr	*46.6*	*5.6*	*0.6*	*0.002*
Pb	*225.4*	*42.1*	*5.2*	*0.01*
Zn	*1316.2*	*326.4*	*36.9*	*0.2*

runoff (20 m^3 $5min^{-1}$) the relationship with TSS was found to be:

$$TSS = -85.47 + 32.28 \ln Q \qquad r^2 = 0.84 \qquad (5)$$

Below 20 m^3 $5min^{-1}$, runoff and TSS exhibited no relationship. The relationship for rising stage is:

$$TSS = 0.22\ Q^{1.25} \qquad r^2 = 0.91 \qquad (6)$$

and for falling stage the relationship is:

$$TSS = -0.13 + 0.46\ Q \qquad r^2 = 0.93 \qquad (7)$$

The loads of heavy metals whether in the soluble or suspended phase are low compared with similar data for the FRG (Reichert & de Haar, 1982). As for TSS, TDS and heavy metal loads, maximum transport rates are related to flood runoff. The heavy metal loads exported from the drainage basin, except for elements such as Pb and Zn, are primarily related to geochemical sources, whereas the storm water loads depend on wet and dry deposition which accumulates between rainfall events.

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