PROTECTION AIMS FOR THE HYDROSPHERE:
CRITICAL LEVELS AND CRITICAL LOADS OF THE NUTRIENTS C, N, P AND S IN SURFACE WATERS AND GROUNDWATER

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Description
Reactive compounds of C, N, P, S cause not only directly eutrophication and acidification of pedosphere and hydrosphere with corresponding decline of biosphere, but also indirectly emissions of acidifiying [i.e. $(CH_3)_2S$] and climate relevant [i.e. CH_4, N_2O[gases **(Fig. 1)**. Hydrosphere comprehends both groundwater and surface waters like the inland waters of running waters and lakes as well as seas / oceans [17].

Rationale mechanism of action
Here primarily the aim is to show as limits critical levels and critical loads for C, N, P, S below these, a good chemical and ecological status of the hydrosphere and its biosphere could be obtained. Those obligatory protection aims (not targets!) determine both the extents and kinds of the preferred mitigation options and measures. Therefore these protection aims for environment are prerequisites for all 8 categories of corresponding cause-oriented and sufficient mitigation options and measures to get a good chemical and ecological status especially here of the hydrosphere [8-18].

Applicability
1. Inconsistency on the one hand between EU Nitrates Directive [CD 91/676 / EC], Water Framework Directive [WFD: 2000/60/ EC] as well as (Draft) Groundwater Directive [KOM 2003, 550 final] and on the other hand to get a good chemical and ecological status of the hydrosphere

1.1 No critical levels of loads for hydrosphere and C, N, P, S are shown in these EU Directives except with the nitrate level of 50 mg $NO_3^{\cdot}$ l^{-1} (11.3 mg NO_3-N$^{\cdot}$ l^{-1}) "to protect the groundwater in such a way that good drinking water can be achieved by simple purification, as specified also in the WFD" [8-10]

1.2 There are no vulnerable zones to (nitrate) leaching as maintained in the EU-Nitrate Directive but only vulnerable (soil use) managements e.g. of the farmers in respect to not tolerable C. N. P. S losses / emissions especially those of agriculture.

1.3 But this limit of 50 mg $NO_3^{\cdot}$ l^{-1} is inconsistent: with human health and the protection of the hydrosphere

1.3.1 Nitrate and human health

a) Nitrate is not involved in methaemoglobinaneamia and stomach cancer **(Tab. 1 / Tab. 2)** [1,4]
b) Nitrate and nitrate levels up to 1000 mg$^{\cdot}$ l^{-1} and also nitrite e.g. in drinking water or beet root juices but also in fruits and vegetables preserve, rather than threatens, health by repressions of:

- Gastroenteritis **(Tab. 1)** [1]
- Dental caries **(Tab. 1**) [1]
- Fungal pathogens on skin **(Tab. 1)** [1]
- Cardiac infection and therefore both nitrate and nitrite serve as essential nutrients for optimal cardiovascular health **(Tab. 3**) [2]
- Too high blood pressure and therefore the risk of cardiovascular disease **(Tab. 4)** [21]

1.2.2 Nitrate and protection of the hydrosphere:

This limit level of 50 mg $NO_3 \cdot l^{-1}$ both in groundwater and surface waters is inconsistent with the protection of the hydrosphere, especially if too high critical loads of nitrate are exported with the running waters to the transition waters (estuaries), coastal waters and to the seas / oceans. Note that the actual “baseline concentrations” e.g. in German main streams are “only” between 1,8 and 6,3 mg NO_3-N $\cdot l^{-1}$ corresponding to 8 and 28 mg $NO_3 \cdot l^{-1}$ [6,17]

2. Critical C, N, P, S levels and loads

2.1 Critical C, N, P, S levels (concentrations)

Tab. 5 shows the qualification of running waters (and groundwater) in respect to the nutrient levels in Germany. Anthropogenically moderately influenced the aim corresponds level II (green), taken not into account downstream protection levels of estuaries, coastal waters and seas / oceans as was done in **Tab. 6**: Here background levels (good – very good) and as possible aimes orientation levels (good – moderate) corresponding about 1.5 background levels of C, N, P in surface waters (and groundwater) in Germany is shown irrespective of the corresponding loads of running waters / rivers and necessary downstream protection especially of estuaries, coastal waters and seas / oceans. Note: The critical level for nitrate in running waters is only < 2.5 mg $\cdot l^{-1}$ compared with the target level of the EU-Nitrate Directive, WFD, Draft Groundwater Directive of 11.3 mg $\cdot l^{-1}$!
Additionally **Tab. 7** shows TP, PO_4-P and Chl-a limits in systems for evaluating the water quality of rivers, lakes and seas [3].

2.2 From critical levels (concentrations) to critical loads in the context of the protection of coastal zones and seas / oceans

Fig. 2 shows the worldwide increase of nitrogen loads in the streams from sources to estuaries, coastal waters and into the seas / oceans. Hot “spots” are primarily in Europe, but also in Asia, North and South America with increase of 400% to more than 500% [19], leading in the coastal zones to nitrogen inputs and surpluses of about 500 (170-900) kg $N \cdot ha^{-1} \cdot yr^{-1}$ and phosphorus inputs and surpluses of about 60 (10-80) kg $P \cdot ha^{-1} \cdot yr^{-1}$ [7]. Up till now measures for reduction of e.g. nitrate losses/emissions from agriculture to groundwater focus only on areas with high nitrate concentrations of > 50 mg $\cdot l^{-1}$ irrespective of the corresponding nitrate loads (e.g. EU-Nitrate Directive, -Water Framework Directive, -Groundwater Directive). In respect to nitrogen emissions to surface waters this strategy will fail! Areas with low concentrations in groundwater tend to be of high importance to nitrogen loads transported towards the seas / oceans, e.g into the Black Sea [22]. Therefore a management strategy for reduction of nitrate emissions only for those areas with high nitrate concentrations in groundwater will fail in respect to protection of surface waters as coastal zones and seas/oceans. Further on the EU-WFD protects not the coastal waters and the seas/oceans, because its range of validity ends 1 sea mile from the coastal line.
Searching for critical loads for running waters especially in respect to downstream protection to the coastal ones and seas / oceans till now the best way is to compare their actual C, N, P (S) loads with those of former times as no environmental problems (e.g. eutrophication) exists. This can be seen e.g. in **Fig. 3** comparing the N and P emissions into the river system of the Danube of 1955 (no eutrophication in the Western Black Sea) with those emissions of 1965-2000 (with eutrophication). [5,8-17]

2.3 No compensation of nutrient emissions reductions between point and diffuse sources, like those of agriculture.
Those compensations as proposed by Trepel and Ollesch 2007 [20] and shown in **Fig. 4** must be refused because e.g. C, N, P, S emissions by point sources as those of wastewater management are only directly into the surface waters. But those of diffuse sources, like of agriculture, are (even mainly in respect to N and S) additionally via the drainage zone and groundwater into the surface waters, with supplementary negative impacts on the drainage and groundwater zones like remobilisation of SO_4^{2-} by NO_3^- or heavy metals, emission of climate relevant gases like N_2O and more or less increasing oxidation processes by NO_3 and SO_4 with all their negative impacts. [17]

Effectiveness, including certainty
The effectiveness and certainty of mitigation options and measures is all the more higher as the differences are between actual and critical C, N, P, S levels and loads.

Time frame
Needed time frames to reach and fall below the critical C, N, P, S levels and loads depends on the kind of sources like point or diffuse sources, time for implementation of nutrient mitigation measures and additional C, N, P, S stocks in soils, drainage and groundwater zones as well as in the sediments of the surface waters in respect on remobilisation processes of C, N, P, S. “Normal” time frames for success can be in respect to point sources within a few years. But in respect to diffuse sources and especially of agriculture success will be in the range of about 20-30 years or even longer with measures like wetland restoration (fens and bogs) or conversion of arable land to grassland of about 30 to 100 years (see factsheet “Soil management/Improve soil organic matter). [17,18]

Environmental side-effects / pollution swapping
There are no negative environmental side-effect from setting critical C, N, P, S levels and loads for the hydrosphere. But in respect to an integrated environmental protection there is a need to fulfil simultaneously also the critical C, N, S levels e.g. for the atmosphere, like those for the eutrophying , acidifying and climate relevant gases e.g. CO_2, CH_4, NMVOC, NH_3 ➔ NH_4^+, N_2O, NO ➔ NO_2, H_2S, SO_2 and fine dust (CPM). Therefore e.g. development of an obligatory best agricultural practice based on those critical C, N, P, S levels and loads is decisive for sufficient and cause-oriented reductions of the C, N, P, S losses and emissions independent of the location of an area. This is including also resource protection esp. in respect to N (fossil energy) and P (fossil mineral P reserves). [8-18]

Relevance, potential for targeting, administrative handling, control
The relevance and potential for setting standards as critical C, N, P, S levels and loads (“targeting”) is extremely high and administrative handling as well as control are prerequisites for their implementation especially here within a catchment river basin scale. [17,18]

Costs, investments, labor
Expenditures for costs, investments and labor are primarily for administrative handling and control in respect to the fulfilment of the critical C, N, P, S levels and loads by the point and diffuse sources, therefore by the total system nutrition with agriculture, human nutrition as well as the waste and wastewater management. But the ecological and social win-win situations will be morefold higher than these expenditures.

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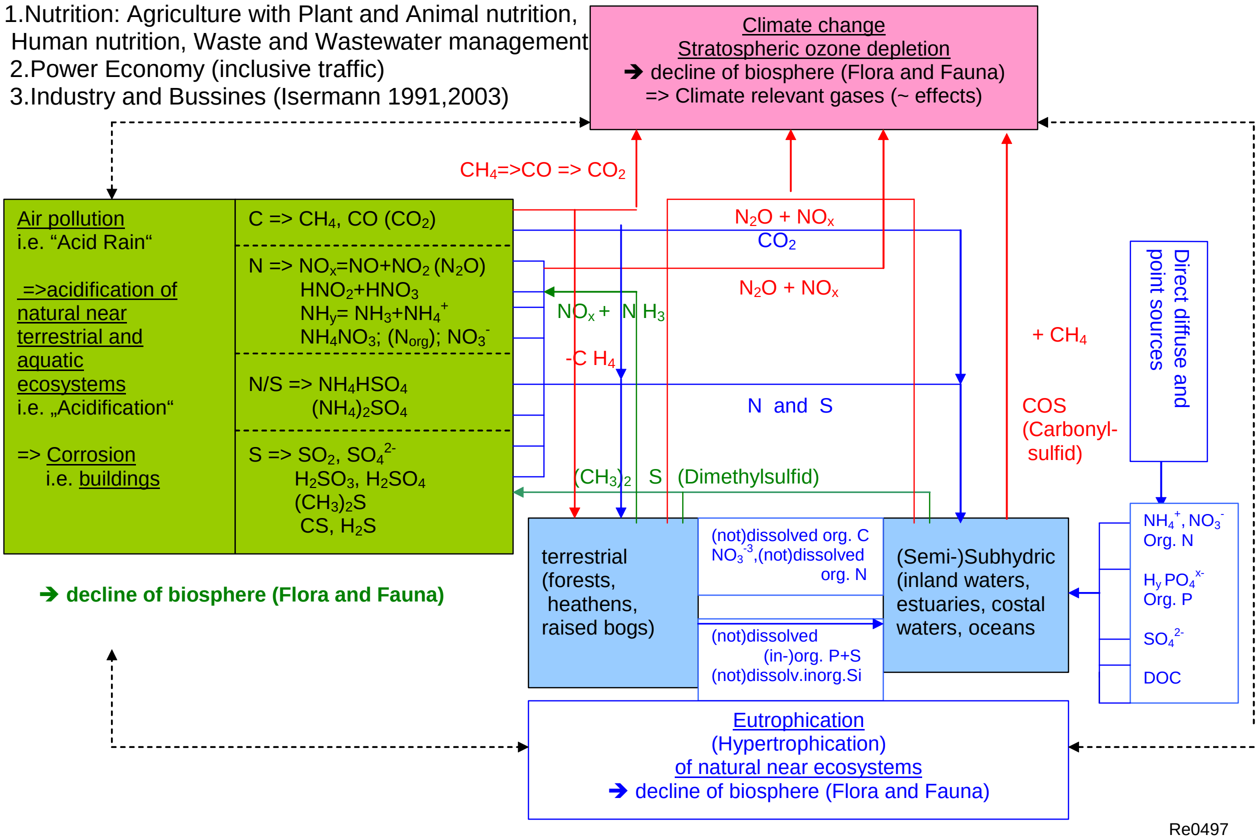
Fig. 1:
Interactions of boundary crossing continental and global environmental damages caused by reactive components of the multifunctional (!) nutrients C, N, S and monofunctional P anthropogenically produced by the sectors:
1.Nutrition: Agriculture with Plant and Animal nutrition, Human nutrition, Waste and Wastewater management
2.Power Economy (inclusive traffic)
3.Industry and Bussines (Isermann 1991,2003)



Tab. 1: Nitrate and human health

[T.M. Addiscott [1)] and N. Benjamin (2004):
Soil Use and Management 20, 98-104]

Abstract

1. **Nitrate is widely and mistakenly perceived to threaten human health by causing methaemoglobinaemia in infants and stomach cancer in adults, but it does cause environmental problems**
   - 1.1 **Methaemoglobinaemia** is a side-effect of gastroenteritis and is not caused by nitrate but by nitric oxide, which is produced I a defensive reaction stimulated by gastroenteritis. The latter may be caused by a bacterium or a virus. The association of methaemoglobinaemia with nitrate may have arisen because early cases of the condition were often associated with wells polluted with bacteria, and the same pollution increased the nitrate concentration.
   - 1.2 Four epidemiological studies sought a link between **stomach cancer** and nitrate but did not find one. The incidence of this cancer has also declined during the last 30 years, while nitrate concentrations in water have increased.
2. **Nitrate preserves, rather than threatens, health**
   - 2.1 It is reduced by microbes on the tongue to nitrite, which generates nitric oxide when acidified in an antibacterial defence mechanism vital to our well-being. This mechanism **acts with great effectiveness in the stomach against *Salmonella, Escherichia coli* and other organism that cause gastroenteri-tis**.
   - 2.2 It also **acts in our mouths against dental caries and even on our skin against fungal pathogens such as *Tinea pedis* (athlete´s foot).** This mechanism is the basis of the centuries-old practice of adding nitrate or nitric to stored meat to protect against botulism, caused by the most lethal toxin know to mankind.

Re0610

Tab. 2: Causes of Cancer and their shares:

- 63% of all cancers are caused by an unhealthy life style
- only 4% by inheritance
- and 0% by nitrate and nitrite

[European Prospective Investigation into Cancer and Nutrition (EPIC-Study 1992-2004)]

Causes of cancer	Shares (%)
1. (Over-)Nutrition	**30**
2. Smoking	**30**
3. Infections	**15**
4. Other factors (like medicine, radiation, immune-suppression, hormones, reproduction-factors)	**13**
5. Professional exposition	**5**
6. Inheritance	**4**
7. Alcohol	**3**
8. Nitrate and nitrite i.e. drinking water or added stored meat [1)]	**0**

[1)] Addiscott and Benjamin (2004): Annex I of D 3.3 re0642

Effects of dietary nitrite and nitrate on myocardial ischemia/reperfusion injury

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Nitrite has emerged as an endogenous signaling molecule with potential therapeutic implications for cardiovascular disease. Steady-state levels of nitrite are derived in part from dietary sources; therefore, we investigated the effects of dietary nitrite and nitrate supplementation and deficiency on NO homeostasis and on the severity of myocardial ischemia/reperfusion (MI/R) injury. Mice fed a standard diet with supplementation of nitrite (50 mg/liter) in their drinking water for 7 days exhibited significantly higher plasma levels of nitrite, exhibited significantly higher heart levels of nitrite, nitroso, and nitrosyl–heme, and displayed a 48% reduction in infarct size (Inf) after MI/R. Supplemental nitrate (1 g/liter) in the drinking water for 7 days also increased blood and tissue NO products and significantly reduced Inf. A time course of ischemia–reperfusion revealed that nitrite was consumed during the ischemic phase, with an increase in nitroso/nitrosyl products in the heart and plasma. Mice fed a diet deficient in nitrite and nitrate for 7 days exhibited significantly diminished plasma and heart levels of nitrite and NO metabolites and a 59% increase in Inf after MI/R. Supplementation of nitrite in the drinking water for 7 days reversed the effects of nitrite deficiency. These data demonstrate the significant influence of dietary nitrite and nitrate intake on the maintenance of steady-state tissue and plasma nitrite/nitroso levels and illustrate the consequences of nitrite deficiency on the pathophysiology of MI/R injury. Therefore, nitrite and nitrate may serve essential nutrients for optimal cardiovascular health and may provide a treatment modality for cardiovascular disease.

re0928

Tab. 3: The repressive role of nitrite and nitrate on cardiac infection

A) Recommended nitrite / nitrate level in drinking water
1. **Nitrite / Nitrate deficiency: 59% increase infarct size**
2. **50 mg nitrite$^-$ l^{-1}: 48% reduction infarct size**
3. **1000 mg nitrate/l (supplemental): significantly reduction infarct size**

B) Threshold values for nitrate in drinking water
1. **Animal nutrition: 300 mg NO_3^- · l^{-1}**
2. **Human nutrition: 40/50 mg NO_3^- · l (? ? ?)**

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Acute Blood Pressure Lowering, Vasoprotective, and Antiplatelet Properties of Dietary Nitrate via Bioconversion to Nitrite

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Abstract—Diets rich in fruits and vegetables reduce blood pressure (BP) and the risk of adverse cardiovascular events. However, the mechanisms of this effect have not been elucidated. Certain vegetables possess a high nitrate content, and we hypothesized that this might represent a source of vasoprotective nitric oxide via bioactivation. In healthy volunteers, approximately 3 hours after ingestion of a dietary nitrate load (beetroot juice 500 mL), BP was substantially reduced (Δ_{max} -10.4/8 mm Hg); an effect that correlated with peak increases in plasma nitrite concentration. The dietary nitrate load also prevented endothelial dysfunction induced by an acute ischemic insult in the human forearm and significantly attenuated ex vivo platelet aggregation in response to collagen and ADP. Interruption of the enterosalivary conversion of nitrate to nitrite (facilitated by bacterial anaerobes situated on the surface of the tongue) prevented the rise in plasma nitrite, blocked the decrease in BP, and abolished the inhibitory effects on platelet aggregation, confirming that these vasoprotective effects were attributable to the activity of nitrite converted from the ingested nitrate. These findings suggest that dietary nitrate underlies the beneficial effects of a vegetable-rich diet and highlights the potential of a "natural" low cost approach for the treatment of cardiovascular disease.

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Key words: diet • nitric oxide • blood pressure • hypertension • ischemia/reperfusion • platelets • endothelium re0975

Tab. 5:Qualification of running waters (= Groundwater/Draft EU-Groundwater Directive 2006) in respect to the nutrient levels C, N, P, S in Germany (LAWA / UBA 2006) (basis value: 90-percentile)

[http:// www. umweltbundesamt.de/wasser/themen/ow_s3_3.htm]

Nutrients	Unit	Chemical water quality categories regarding the nutrient levels, irrespectively of their loads						
		I	I-II	II[1)]	II-III	III	III-IV	IV
		Anthropogenic Levels						
		Not influenced	very low	moderate	obvious	increased	high	very high
		Geogenic background	½ aim	aim	up to 2x aim	up to 4x aim	up to 8x aim	up > 8x aim
		dark-blue	light-blue	green	light-green	yellow	orange	red
1. TOC	$mg \cdot l^{-1}$	<= 2	<=3	**<= 5**	<= 10	<= 20	<= 40	> 40
2. total N(TN)	$mg \cdot l^{-1}$	<=1	<= 1,5	**<= 3**	<= 6	<= 12	<= 24	>24
…of it:								
2.1 Nitrate-N	$mg \cdot l^{-1}$	<= 1	<= 1,5	**<= 2,5[2)] (drinking water:<11,3)**	<= 5	<= 10 (drinking water:<11,3)	<= 20	>20
2.2 Nitrite-N	$mg \cdot l^{-1}$	<= 0,01	<= 0,05	**<= 0,1**	<= 0,2	<= 0,4	<= 0,8	>0,8
2.3 Ammonium-N	$mg \cdot l^{-1}$	<= 0,04	<= 0,1	**<= 0,3**	<= 0,6	<= 1,2	<= 2,4	> 2,4
3. Total-P(TP)	$mg \cdot l^{-1}$	<= 0,05	<= 0,08	**<= 0,15**	<= 0,3	<= 0,6	<= 1,2	> 1,2
…of it: Ortho-Phosphate-P	$mg \cdot l^{-1}$	<= 0,02	<= 0,04	**<= 0,1**	<= 0,2	<= 0,4	<= 0,8	> 0,8
4. Sulfate	$mg \cdot l^{-1}$	<= 25	<= 50	**<= 100**	<= 200 (drinking water: = 240)	<= 400	<= 800	> 800

1) Demand up to 2010: strict observance on all measuring locations of LAWA

2) **Maximum tolerable for total life of adults (70 kg life weight, 2 l drinking water $\cdot$ d^{-1}): 2,9 mg NO_3-N $\cdot$ l^{-1}**

Re0784

Tab. 6: Background levels (BG: good-very good) and as possible aims orientation levels (OR: good-moderate) of the nutrients C, N, Pin surface waters (and groundwater) in Germany irrespective of the corresponding loads of running waters/rivers and necessary downstream protection especially of transition waters (estuaries) and coastal waters according to:
A) LAWA-AO-Rahmen Konzeption (RAKON /07.03.2007) in comparison with B) LAWA-AO-Rahmen Konzeption (RAKON /07.03.2007)
(Isermann / BSNLC 2007)

Qualifica-tions	A) LAWA-AO-Rahmen Konzeption (RAKON /07.03.2007) (http://www.wasserblick.net/servlet/is/42489/?lang=de&highlight=rakon)							B) LAWA /EPA-Germany 2005/2006 (http://www.umweltbundesamt.de/ wasser/themen/ow_s3_3.htm)		
Surface waters	LAKES [Vegetation averages]	RUNNING WATERS [averages]		TRANSITION AND COASTAL WATERS: Running waters flowing into coastal waters with retention (➔Remobilisation?) Calculated with MONERIS				SURFACE WATERS (AND GROUNDWATER) [3)] [90-percentile]		
				North Sea resp. ( ) German Bight		Baltic Sea resp. ( ) Arkona Sea				
Nutrient levels	BG	BG	OR	BG	OR =1.5 BG	BG	OR =1.5 BG	BG (I)	½ Aim (I /II)	Aim (II) (moderate)
TOC (mg/l)	-	5 -10	7 - 15	-	-	-	-	← 2	← 3	← 5
TN[1)] (mg/l)	-	in preparation	in preparation	0.21-0.35 (0.17) [year]	0.42-0.70 (0.34) [year]	0.14 (0.14) [year]	0.21 (0.21) [year]	← 1	← 1.5	← 3
DIN (mg/l)	-	-	-	0.13-0.24 (0.13) [winter]	0.26-0.48 (0.26) [winter]	0.08-0.11 (0.030-0.035) [winter]	0.15-0.17 (0.040-0.050) [winter]	-	-	-
NO_3-N (mg/l)	-	in preparation	in preparation	0.10-0.18 (0.090) [winter]	0.20-0.36 (0.180) [winter]	0.04-0.07 (0.030-0.035) [winter]	0.06-0.11 (0.040 -0.050) [winter]	← 1	← 1.5	← 2,5
NO_2-N (mg/l)	-	-	-	-	-	-	-	← 0.01	← 0.05	← 0.10
NH_4-N (mg/l)	-	0.02-0.04	0.10 – 0.30	-	-	-	-	← 0.04	← 0.10	← 0.30
TP[2)] (µg/l)	6-86	50-100	100-300	10-30 (20) [year]	20-50 (40) [year]	9-19 (14) [year]	25-28 (28) [year]	← 50	← 80	← 150
PO_4-P (µg/l)	-	10-20	70 -200	4-8 (8) [winter]	8-16 (16) [winter]	4-18 (8-9) [winter]	12-24 (12-14) [winter]	← 20	← 40	← 100

1) TN: 1µ mol/l= 0.014 mg/l; 2) TP: 1µ mol = 0.031 mg/l

3) **The level of protection against new discharges, emissions and losses must be at least comparable to that for surface water of good chemical status (Amendment 3, Recital 1a, European Parliament Legislative Resolution on the Council ➔ Draft EU-Groundwater Directive, June 13, 2006)**

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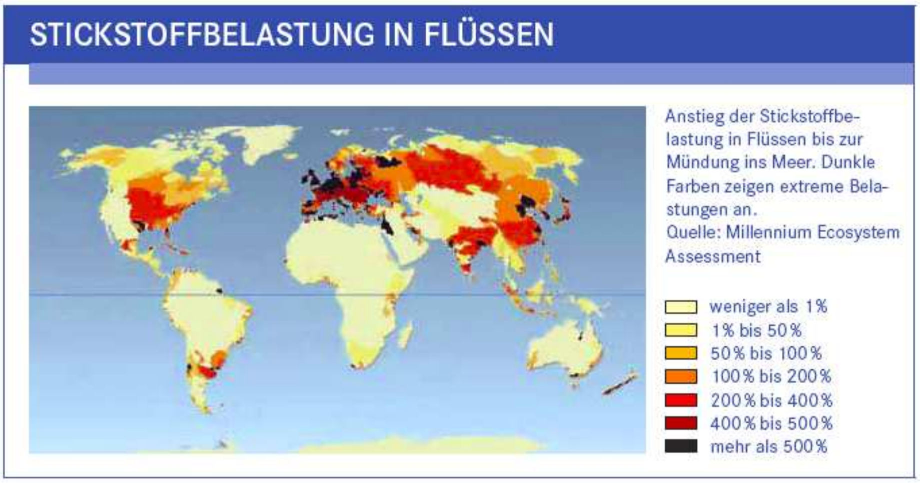
Tab. 7: TP, PO_4-P and Chl-a limits in systems for evaluating the water quality of rivers, lakes and seas

(Csathó et al. 2007)

Water bodies	Measured properties	Units	Quality classes (I is the best)				
			I	II	III	IV	V
Rivers	TP	(mg l^{-1})	<0.1	0.1-0.2	0.2-0.4	0.4-1	>1
	PO_4-P	(mg l^{-1})	<0.05	0.05-0.1	0.1-0.2	0.2-0.5	>0.5
	Chl-a	(µg l^{-1})	<25	25-50	50-100	100-250	>250
Lakes	TP	(mg l^{-1})	<0.005	0.005-0.01	0.01-0.03	0.03-0.1	>0.1
	PO_4-P	(mg l^{-1})	-	-	-	-	-
	Chl-a	(µg l^{-1})	<2.5	2.5-8	8-25	25-75	>75
[a]Seas	TP (summer)	(mg l^{-1})	<0.0145	0.0145-0.0231	0.0231-0.0386	0.0386-0.1157	>0.1157
	PO_4-P (winter)	(mg l^{-1})	<0.0125	0.0125-0.02	0.02-0.0333	0.0333-0.1	>0.1
	Chl-a	(µg l^{-1})	<1.29	1.29-2.06	2.06-3.43	3.43-10.29	>10.29

a: based on the reference conditions and 50 % acceptable deviation given in HELCOM (2005c)

re0850



re0974

Fig. 2: Nitrogen loading of streams in the world: Increase of nitrogen loads in the streams from sources to estuaries and coastal waters into the seas / oceans in the range of lower than 1% (light-yellow) to more than 500% (black) [Millenium Ecosystmes Assessment 2008]

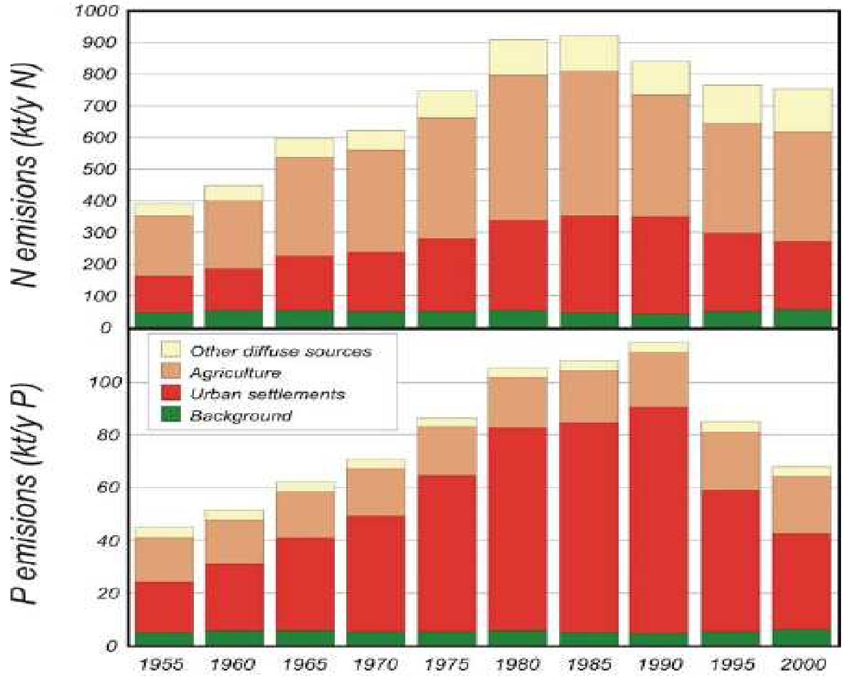


Fig.3: Changes of nitrogen and phosphorus emissions into the river system of the Danube from 1955 to 2000 (Behrendt 2003) re0786

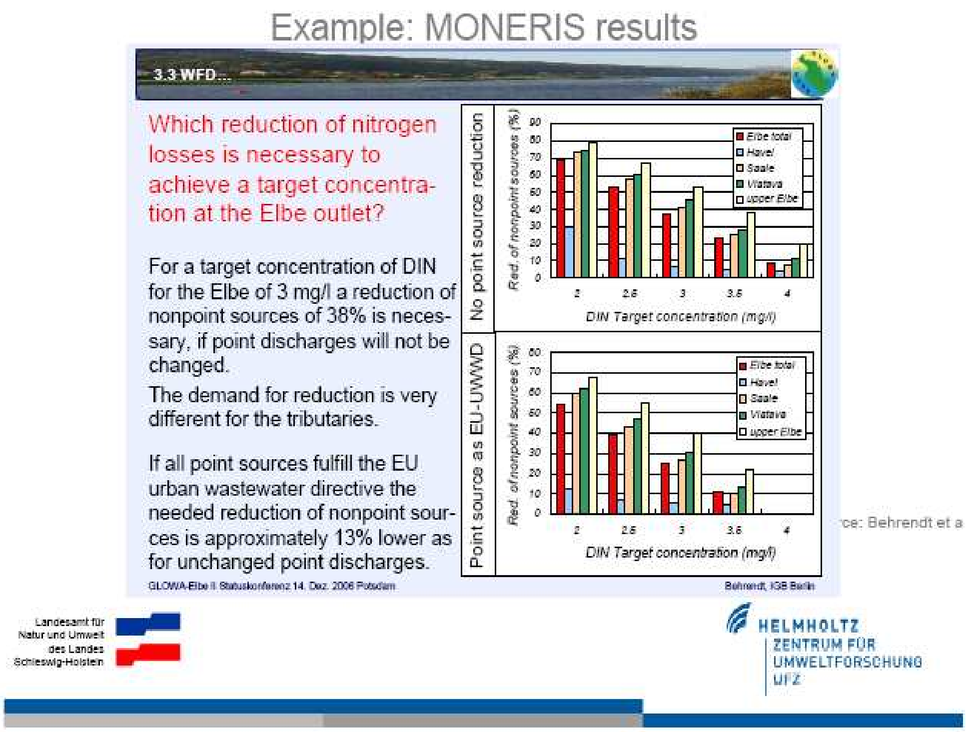


Fig. 4: No compensation of nutrient emission reductions between point and diffuse sources like agriculture as proposed by Trepel and Ollesch (2007) re0939