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ANALYSIS OF THE METHOD FOR DETERMINING THE LOAD OF NUTRIENTS DISCHARGED FROM THE DRAINAGE AREA TO SURFACE WATER**

The purpose of the paper was to compare the quantities of annual loads of nutrients running off the experimental catchment area, calculated by different methods . Results of calculations practically consistent with the real quantity, i.e., calculated from the everyday analyses, have been obtained when the samples were taken everyday and the samples taken during 7–10 days were next poured together proportionally to the flow rate (after being fixed with $HgCl_2$). The most reliable results were obtained from the relation L versus Q by applying a constant sampling rhytm at at least 36 samples a year. Other calculation methods, based on irregular samplings, were characterized by a high randomness of results expressed in remarkable divergence and sometimes high errors observed in particular in the case of phosphorus.

1. INTRODUCTION

The loads of nutrients of nitrogen and phosphorus compounds in particular are a serious hazard for surface water, especially for the stagnant ones. The more so for this type of water since limit loads of nitrogen and phosphorus are in general so low that even the elimination of the input of those elements from the point sources does not protect the lakes and reservoirs against the consequences of eutrophication processes, the intensification of which is very often due to the increasing inflow of nutrient from the non-point sources exclusively. Hence, an intense development of the initiatied in sixties investigations aimed at determining the coefficients of the outflow of nutrients from the drainage areas characterized by different way and degree of their management.

As a rule a correct determining of the quantities of non-point pollutions as well as

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estabilishing of univocal indices of their run-offs are related to serious methodical and technical difficulties. The difficulties in interpretation are chiefly due to the fact that the quantity of the outflowing non-point pollutant loads depends on the whole complex of factors and that a decisive role may be performed by different factors, as a rule, not single ones but in different configurations, depending on the local and actual conditions. Therefore very often both the results and conclusions of many authors are not confirmed by investigations carried out in other regions, and characteristic indices of run-off established by various authors show significant differences and discrepancies. The latter are also undoubtly due to the different methods of investigations and their interpretations. Hence very often results obtained by various authors cannot be compared or generalized. VOLLENWEIDER and DILLON [18] stated that in order to advance our recognition of the problem of water pollution with phosphorus compounds, some studies must be performed, among others on the improvement of measuring methods and ways of determining the run-off factors for catchment basins used in different ways, especially for the agricultural and urban purposes.

Unit indices of the run-off of the substance being analysed are determined by the quantity of non-point pollutants outflowing from the catchment basin. To this end both the annual quantity of outflowing loads and the area of the given catchment basin must be known.

The methods and ways of determining the quantities of loads applied by various authors are different. The following ones are most often found in the literature:

1. The averaged concentrations and flow rates determined in different investigations are multiplied; the obtained product multiplied by 365 yields the annual load [1], [3], [15].

2. It is assumed that for the days between examinations the load (or concentration) and flow rate determined in a given day are constant quantities; their sum yields the annual load [3], [8], [9].

3. The concentration stated in a given day, assumed constant for the days between examinations, is multiplied by flow rate measured everyday [21].

4. Concentration determined from the samples taken continuously and proportionally to the flow rate or time is multiplied by the sum of flow rates [16], [17].

5. The concentration determined either everyday or from the linear interpolation for the days between examinations is multiplied by the flow rate recorded everyday [21].

6. The correlation between concentration and flow rate determined from the random examinations is a basis for determining daily loads, provided that the flow rates are known [7], [12], [17], [20], [11], [10].

To make the interpretation easier, and sometimes even possible, as well as to determine exactly the loads, some authors introduce the notion of characteristic or periodical loads, i.e., of loads established separately for characteristic periods, e.g., for seasons [10], [5].

According to WEBER [21], who compared the results obtained by the methods 2, 3, and 5, the methods 2 and 5 are the least and the most accurate, respectively, and the methods 3 and 5 give similar results if the time intervals between the separate measurements are sufficiently small. Of the methods 1, 2, 4, and 6 KRUMMENACHER [10] considers the method 6 as the most accurate one, but in case of the lack of some relationship between Q and c he decided to resign from it and to apply the method 4.

The sampling frequency and the number of results indispensable for determining correctly the annual load is a controversial problem. WEISNER and LINK [22] took samples everyday; BERNHARDT, SUCH, and WILHELM [3] took samples once a week stating that the obtained values are only tentative and that the analyses should be performed 3 times a week; KRUMMENACHER [15] took samples every 2–3 week, but Q was continuously recorded by means of a limnigraph; AHLGREN [1], KLIFFMUELLER [9], NÜMANN [13], and ODERMATT [14] took analyses once a month. According to WAGNER [19] and UNGER [17] annual loads can be determined correctly if the flow rates are measured within their whole ranges, water samplings taken at constant, possibly small time intervals being not so essential. UNGER [17] is of the opinion that 11 results obtained during 1 year are sufficient to determine annual loads with the desired accuracy, but then all the flow rates must be recorded because of their high contribution to the total load. AMBÜHL [2], who took samples once a week from the Hallwilersee tributaries, states that the extreme situations, almost never recorded by him, are of small importance.

Studies on the evaluation of loads within the total area are very toilsome, laborious and require though different but always high expenditures, depending on the method applied. From the review of the most often used methods and opinions represented by different authors it follows that conclusions concerning the reliability of results obtained by employing different methods and different sampling frequencies have not yet been univocally specified. Hence, the studies which may contribute to solving this problem and above all the search of methods which would give the results most resembling the real conditions at very low expenditures, i.e., the search of the most efficient methods, are essential for standardization and rationalization of the investigations and for obtaining reliable results.

The purpose of the present paper was to establish the method giving the most adequate results of the research works, the range and technique of which would require the least possible expenditures.

2. METHODS

The presented study is based on the data set and results of one-year investigations performed in the catchment basin of the stream Młynarka. This stream is a tributary to the river Bystrzyca. Its catchment area (15.2 km^2) situated in the territory of Sowie Mountains (Lower Silesia) is characterized by the average falling gradient of 31% density of the river system of 0.94% m/km^2. Arable land takes 48% (crop land 22.4%) and forests 45% of the total area. Total loading of the catchment area in the period of investigations amounted to 1.1 t P/km^2·yr and 7.5 t N/km^2·yr (fertilizers, precipitation, population, livestock). Everyday water samplings with simultaneous measurements of the flow rates within the profile closing the catchment area were performed since 1st October 1981 to 30th December 1982. Total phosphorus (TP), total nitrogen (TN), and mineral nitogen ($NH_4 + NO_2 + NO_3 = MN$) in water samples were determined everyday, At the same time other water samples taken and poured together proportionally to the flow rate made averaged samples (fixed with $HgCl_2$) for periods of 7 to 10 days. In these samples TP, TN, and MN were determined like in samples taken everyday. In this way, after a one-year cycle of investigations the obtained data set consisted of 365 results of everyday measurements of flow rate, 365 concentrations of TP, TN, and MN determined everyday, and 51 results obtained from the averaged periodical samples. During the investigations flow rates ranged from 0.35 m^3/s to 1.40 m^3/s, and the mean monthly flow rates, from 0.037 m^3/s to 0.255 m^3/s.

This data set was used for analysis of the reliability of annual loads (L_y) determined by two methods. It has been assumed that the quantity L_y calculated as a sum of products (obtained from multiplication of concentrations determined everyday by the corresponding daily flow rates) is a real quantity L_r and constitutes the reference level for the results obtained by the application of the following two methods:

METHOD I (MI). This method based on periodic samples taken proportionally to the flow rate; L_y was calculated as a sum of products given by multiplication of the concentration determined in each sample by global flow rate in the period this sample was taken.

METHOD II (MII). Calculation of L_y was based on the relationship between the quantity of instaneous load L and flow rate Q at different rate and frequency of sampling. The purpose of this method was to determine the optimal number of observations in order to obtain a correlation between L and Q which, when applied to calculations of L_y, would yield results the closest possible to L_r. In this method two variants of procedure were used:

VARIANT A (VA). It consisted in simulated research (samplings and measurements of flow rates) performed at constant dates (days of the week) and at the following frequency (during one year): 24 analyses (first and third Mondays of each month), 36 analyses (first, second, and third Mondays of each month), 52 analyses (all the Mondays in the year), 105 analyses (all the Mondays and Thurstdays in the year), and 365 daily analyses.

Thus, in this variant the correlation between L and Q used for determining the quantity of everyday loads was based on the 24, 36 or 105 analyses performed regularly or on everyday analyses.

VARIANT B (VB). The following numbers of analyses during one year were assumed in this variant: 24, 36, 48 and 108. The dates were determined by means of computer, applying a generator of pseudorandom numbers from a uniform distribution on (0, 1) interval. For the assumed 24 analyses the dates were chosen in 16–17 series and for the remaining ones, in 12 series.

The quantities of the annual loadings of the investigated nutrients were established by applying the programme LAMANA (DĄBROWSKI, KAPŁAŃSKA [4]) based on the assumptions made by the authoress. The programme expresses a regressive relationship between flow and load. In order to determine the relationship between the flow x and the load y the following family of broken lines was assumed:

$$\begin{aligned} y &= Dx && \text{for } x < x_0, \\ y &= Ax+B && \text{for } x \geqslant x_0. \end{aligned} \tag{1}$$

The broken regression line *1* is identical with the regression line for the pair of points (x_1,y_1), $(x_2,y_2),\ldots,(x_n, y_n)$; in the point x_0, equal to the smallest observed value of x (flow), it bends so as to connect two points (0, 0) and (x_0, y_0) where $y_0 = Ax_0+B$. In the case when the broken line passes below the x-axis ($D < 0$ or $x_0 = 0$) we apply the following model

$$y = AG\cdot x \tag{2}$$

which is optimal straight line passing through (0, 0) and interpolating the system of points (x_1,y_1), $(x_2, y_2),\ldots,(x_n, y_n)$.

3. RESULTS AND DISCUSSION

Annual loads L_y of total phosphorus, total nitrogen, and mineral nitrogen obtained by the methods mentioned above are presented in the table. This table contains also the coefficients of correlations for the equations describing the relationship between L and Q and the calculation of error of L_y with respect to L_r (given in per cent).

From the data obtained in follows that L_y calculated from the averaged periodical samples taken proportionally to the flow rates (MI) is for TP equal to L_r and the error for MN and TN does not exceed 1.2%. Results obtained by this method may be assumed also for nitrogen as being very close to the real ones.

When the samplings and measurements of flow rates are performed regularly at a constant rhythm, then in order to establish the correlation of L and Q, being the basis for reconstruction of the everyday loads (MII, VA) for the calculated L_v (TP), it should be taken into account that the error may increase: from 7.8% at 105 analyses during the year to +26.4% at 24 analyses; for MN the error of the computed L_y ranges from −4.6% to +2.4%, and for TN it varies from −2.8% to +1.1%. The

Table

Quantities of annual loads calculated from the correlation relationships L versus Q by applying various methods and different numbers of examinations

Methods and variant of calculations	Numbers of examinations	TP(1)				MN(2)			TN(3)		
		Order of drawing lots	$L_y^{(4)}$ kg	$r(x,y)^{(5)}$	Error %	$L_y^{(4)}$ kg	$r(x,y)^{(5)}$	Error %	$L_y^{(4)}$ kg	$r(x,y)^{(5)}$	Error %
1	2	3	4	5	6	7	8	9	10	11	12
$L_r^{(6)}$			337.9	–	–	5 285.5	–	–	6 705.6	–	–
MI			337.8	–	0.0	5 349.4	–	+1.2	6 777.3	–	+1.1
M II VA	24		427.1	0.799	+26.4	5 043.9	0.795	−4.6	6 614.7	0.864	−1.4
	36		368.2	0.682	+9.0	5 144.0	0.773	−2.7	6 518.0	0.834	−2.8
	52		307.8	0.923	−8.9	5 410.2	0.995	+2.4	6 663.3	0.996	−0.6
	105		311.6	0.873	−7.8	5 350.3	0.990	+1.2	6 668.4	0.992	−0.5
	365		337.9	0.702	0.0	5 285.4	0.981	0.0	6 705.0	0.982	0.0
M II VB	24	1	331.9	0.961	−1.8	5 185.0	0.999	−1.9	6 558.0	0.999	−2.2
		2	507.8	0.663	+50.3	5 703.0	0.986	+7.9	7 281.0	0.980	+8.6
		3	269.0	0.882	−20.4	5 829.0	0.956	+10.3	7 176.0	0.966	+7.0
		4	291.3	0.980	−13.8	5 670.0	0.908	+7.3	7 036.0	0.922	+4.9
		5	274.9	0.078	−18.7	4 570.0	0.970	−13.5	6 054.0	0.980	−9.7
		6	319.8	0.927	−5.4	4 885.0	0.993	−7.6	6 145.0	0.610	−8.4
		7	372.6	0.789	+10.3	4 894.0	0.979	−7.4	6 331.0	0.966	−5.6
		8	379.6	0.680	+12.3	5 582.0	0.854	+5.6	7 087.0	0.953	+5.7
		9	480.1	0.696	+42.1	5 260.0	0.835	−0.5	7 016.0	0.890	+4.6
		10	339.3	0.651	+0.4	5 138.0	0.999	−2.8	6 862.0	0.989	+2.3
		11	298.2	0.980	−11.8	4 949.0	0.803	−6.4	6 557.0	0.857	−2.2
		12	297.4	0.975	−12.0	5 584.0	0.993	+5.6	6 914.0	0.995	+3.1
		13	999.1	0.989	+195.6	5 120.0	0.996	−3.1	6 308.0	0.996	−5.9
		14	317.1	0.746	−6.2	4 898.0	0.993	−7.3	6 781.0	0.995	−1.1
		15	347.1	0.766	+2.7	4 590.0	0.788	−13.2	6 107.0	0.722	−8.9
		16	295.9	0.708	−12.4	4 348.0	0.813	−17.7	6 318.0	0.824	−5.8
		17	–	–	–	4 654.0	0.639	−11.8	5 739.0	0.848	−14.4

1	2	3	4	5	6	7	8	9	10	11	12
	36		292.0	0.547	−13.3	5 273.0	0.880	−0.2	6 429.0	0.907	−4.1
			304.5	0.457	−9.8	4 745.0	0.854	−10.2	6 600.0	0.911	−0.2
			1053.0	0.974	+211.5	5 562.0	0.868	+5.2	7 450.0	0.988	+11.1
			321.4	0.676	−4.9	5 174.0	0.753	−2.1	6 639.0	0.804	−1.0
			286.5	0.979	−15.2	5 453.0	0.998	+3.2	6 757.0	0.998	+0.8
			343.5	0.653	+1.6	5 118.0	0.773	−3.2	7 015.0	0.844	+4.6
			383.2	0.855	+13.4	4 919.0	0.866	−6.9	6 422.0	0.902	−4.2
			273.5	0.763	−19.1	5 650.0	0.953	+6.9	6 996.0	0.964	+4.3
			332.0	0.950	−1.7	5 242.0	0.992	−0.8	6 708.0	0.993	0.0
			416.6	0.627	+23.3	5 221.0	0.922	−1.2	6 779.0	0.921	+1.1
			439.1	0.632	+28.9	5 037.0	0.929	−4.7	6 599.0	0.924	−1.6
			251.4	0.167	−25.6	4 749.0	0.754	−10.1	6 066.0	0.742	−9.5
	48		340.5	0.928	+0.8	5 653.0	0.987	+6.9	7 051.0	0.985	+5.2
			829.2	0.965	+145.4	5 051.0	0.972	−4.4	7 207.0	0.983	+7.5
			288.0	0.940	−14.8	5 499.0	0.995	+4.0	6 813.0	0.995	+1.6
			341.5	0.518	+1.1	4 991.0	0.735	−5.6	6 520.0	0.759	−2.8
			395.5	0.684	+17.0	5 115.0	0.997	−3.2	6 550.0	0.995	−2.3
			754.0	0.910	+123.1	5 438.0	0.964	+2.9	7 332.0	0.979	+9.3
			315.0	0.961	−6.8	5 145.0	0.996	−2.7	6 602.0	0.997	−1.5
			302.6	0.941	−10.4	5 379.0	0.995	+1.8	6 791.0	0.997	+1.3
			310.3	0.968	−8.2	5 122.0	0.998	+3.1	6 414.0	0.946	−4.3
			411.6	0.690	+21.8	4 845.0	0.965	−8.3	6 486.0	0.949	−3.3
			393.2	0.709	+16.3	5 334.0	0.992	+0.9	6 816.0	0.985	+1.6
			275.1	0.929	−18.6	4 673.0	0.979	−11.6	5 995.0	0.984	−10.6
	108	1	356.3	0.624	+5.5	5 399.0	0.983	+2.1	6 787.0	0.979	+1.2
		2	553.8	0.805	+63.9	5 332.0	0.967	+0.9	7 024.0	0.978	+4.8
		3	367.5	0.665	+8.7	5 336.0	0.991	+0.9	6 747.0	0.986	+0.6
		4	389.8	0.669	+15.3	5 305.0	0.978	+0.4	6 850.0	0.980	+2.1
		5	356.3	0.672	+5.4	5 367.0	0.978	+1.5	6 874.0	0.977	+2.5
		6	298.9	0.961	−11.6	5 216.0	0.980	−1.3	6 474.0	0.986	−3.4
		7	293.6	0.969	−13.1	5 323.0	0.996	+0.7	6 644.0	0.995	−0.9
		8	288.6	0.928	−14.6	5 143.0	0.965	−2.7	6 492.0	0.976	−3.2
		9	348.4	0.717	+3.1	5 371.0	0.993	+1.6	6 805.0	0.992	+1.5
		10	314.5	0.904	−6.9	5 199.0	0.961	−1.6	6 498.0	0.970	−3.1
		11	592.8	0.846	+75.4	5 568.0	0.937	+5.3	7 284.0	0.954	+8.6
		12	325.5	0.889	−3.7	5 408.0	0.975	+2.3	6 880.0	0.979	+2.6

values of L_y, computed for TP, MN, and TN based on the equation derived from 365 analyses during one year, are consistent with L_r.

In 16 series of random choice of dates (MII, VB) at 24 dates in the year the error of calculated L_y (TP) ranged from -20.4% to $+195.6\%$, in 17 series of the choice of L_y (MN) it varied from -17.7% to $+10.3\%$, while for L_y(TN) it ranged within -14.4% and $+8.6\%$. A significant reduction of the range of errors, though still too large for the reliability of the obtained results (especially for L_y (TP)), has been stated at 108 analyses in the year, where for L_y (TP) it oscillated from -14.6% to $+75.4\%$, for L_y (MN), from -2.7% to -5.3%, and for L_y (TN), from -3.4% to -8.6%.

The presented results give the evidence to a high randomness of correlations between L(TP) and Q which is the higher the smaller is the number of the data used to its determining. This statement is illustrated graphically by a remarkable dispersion and divergent course of lines determining the dependence of L_y (TP) versus Q at different numbers of data (figs. 1–3); it is also confirmed by the data presented in the table. According to them the value of L_y (TP) computed from the regression equations determined from, e.g., 24 random analyses (MII, VB) for which the correlation coefficients $r(x, y) > 0.980$, may exceed L_r by 195.6% or equally well be by 13.8% lower. At 36 random analyses the regression equations with the correlation coefficients $r(x, y) > 0.970$ lead to the values of L_y (TP) by 211.5% higher or 15.2% lower than L_r.

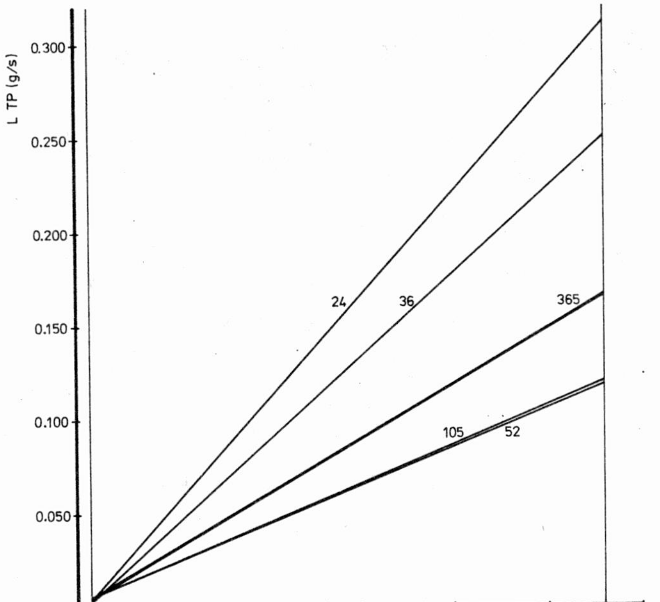

Fig. 1. Correlation relations between total phosphorus load L(TP) and flow rate Q at a constant rhythm of samplings, based on different numbers of analyses (24, 36, 52, 105, 365)

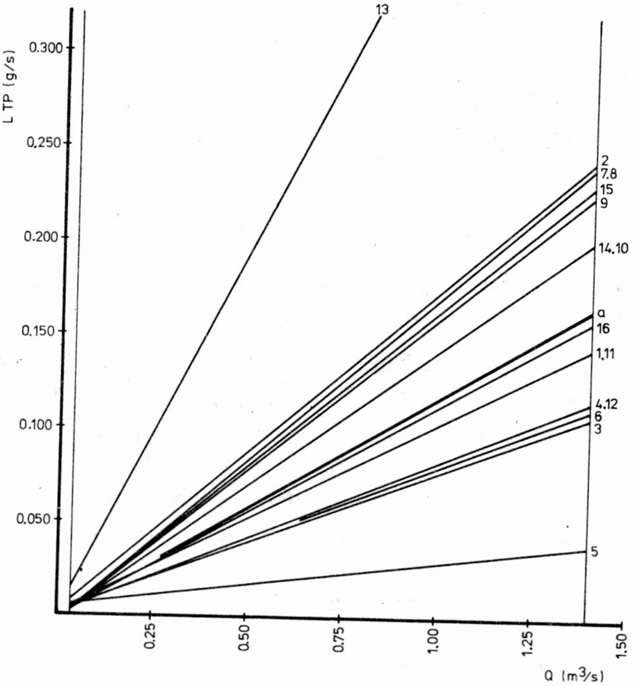

Fig. 2. Correlation relations between the total phosphorus load L(TP) and flow rate Q determined for 24 analyses in 16 random series

a – L versus Q determined from 365 analyses, 1–16 consecutive numbers in a series

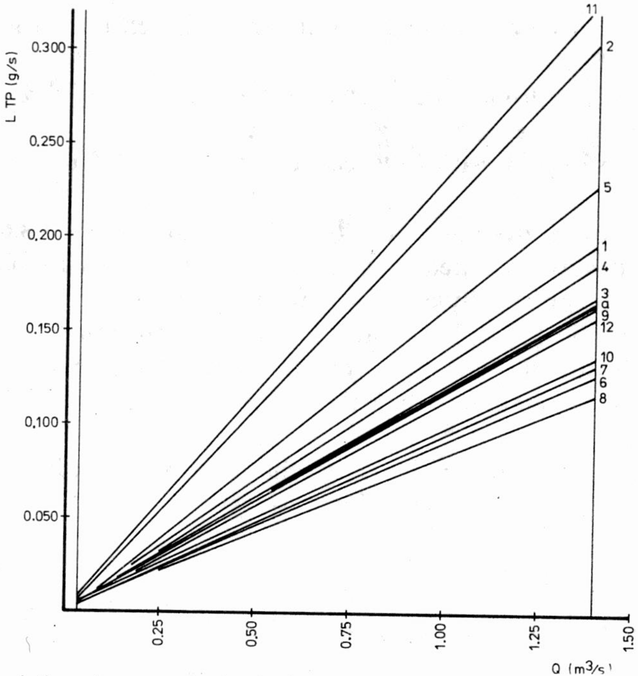

Fig. 3. Correlation relations between the load of total phosphorus L(TP) and flow rate Q determined for 108 analyses in 17 series

a – L versus Q determined from 365 analyses, 1–17 consecutive numbers in a series

4. CONCLUSIONS

1. Real quantity of the annual load of nutrients may be determined from water samples taken and analysed everyday and the flow rate being simultaneously recorded. This method requires 365 analyses performed during one year.

2. An identical value for the total phosphorus and for mineral and total nitrogen charged with 1% error may be obtained by taking everyday samples of water and fixing them with $HgCl_2$ and then by making samples for the period of 7 to 10 days, pouring them together proportionally to the flow rates recorded everyday. This method reduces the number of indispensable analyses to 52–37 in one year.

3. When a constant rhythm of taking momentary samples is applied, the flow rate being simultaneously measured and the quantities of loads for the days between the samplings are reconstructed from the relationship betwen L and Q, then the possibility of the following errors should be taken into account:

a) at 24 data (sampling two times a month) the calculation errors for the annual load of total phosphorus, mineral nitrogen, and total nitrogen reach +26.4%, −4.6%, and − 1.4%, respectively,

b) when the number of data increases from 36 to 105, the error decreases and ranges from +9% to −7.8% for total phosphorus, from +2.4% to −2.7% for mineral nitrogen, and from −2.8% to 0.5% for total nitrogen,

c) at 365 data taken for determining L versus Q the results obtained are consistent with the real load, thus giving the evidence to the correctness of the programme LAMANA.

4. When the sampling dates are taken at random, then the error in the applied series ranged as follows:

a) for annual load of total phosphorus it ranged from +145.4% to +211.5% at 24 to 48 data and decreased to 75% at 108 data,

b) for mineral nitrogen it varied respectively from +10.3% to −17.7% and from +5.3% to −2.7%,

c) for total nitrogen, from +11.1% to −14.1% and from +8.6% to −3.4%.

Summing up, it may be stated that the result of the calculations of the annual load, which would be practically consistent with the real value, is obtained by using the method of periodical samples poured together proportionally to the flow rate.

The result only slightly deviating from the real value is obtained by applying a constant rhythm of sampling when the number of data is not smaller than 36 a year.

The remaining calculation methods are characterized by a high randomness of results which may be charged by serious errors.

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ANALIZA METOD OBLICZANIA ŁADUNKU BIOGENÓW WYMYWANYCH ZE ZLEWNI DO WÓD POWIERZCHNIOWYCH

Porównano wielkości rocznych ładunków związków biogennych, odpływających ze zlewni doświadczalnej, obliczonych różnymi metodami. Wynik obliczeń praktycznie zgodny z wielkością rzeczywistą, tj. obliczoną na podstawie codziennych badań, uzyskano zlewając próbki pobrane w ciągu 7 do 10 dni proporcjonalnie do wielkości przepływu. Próbki były utrwalane $HgCl_2$. Wyniki najmniej odbiegające od wielkości rzeczywistych otrzymano ze związku pomiędzy L a Q stosując stały rytm poboru próbek, dający

nie mniej niż 36 danych w roku. Inne metody obliczeń przy nieregularnym poborze próbek cechowała duża przypadkowość wyników wyrażająca się znaczną rozbieżnością i niekiedy dużymi, szczególnie w przypadku fosforu, błędami.

АНАЛИЗ МЕТОДОВ ВЫЧИСЛЕНИЯ ЗАРЯДА БИОГЕНОВ, ВЫМЫВАЕМЫХ ИЗ БАССЕЙНОВ К ПОВЕРХНОСТНЫМ ВОДАМ

Сравнены величины годовых зарядов биогенных соединений, отливающихся из экспериментального бассейна, вычисленных разными методами. Результат вычислений, практически согласный реальной величине, т.е. вычисленный на основе ежедневных исследований, получили, сливая пробы, взятые в течение 7–10 дней пропорционально величи её течения. Пробы были закрепляемы $HgCl_2$. Результаты, наименее отходящие от реальных величин, получены из связи L и Q, применяя постоянный ритм отбора проб, дающий не менее, чем 36 данных в год. Другие методы вычислений при нерегулярном отборе проб характеризовала большая случайность результатов, выражена значительным расхождением и время от времени большими, особенно в случае фосфора, ошибками.