LONG-TERM TRENDS IN THE CHEMICAL AND MECHANICAL DENUDATION IN A SMALL CARPATHIAN CATCHMENT

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Abstract: Chemical and mechanical denudation are among the most important morphogenetic processes, especially in mountainous areas. The main objective of this study was to calculate the chemical deposition in precipitation as well as the chemical and mechanical deposition in a Carpathian stream between 1995 and 2023. The chemical denudation balance was also determined. Concentrations of several ions, including Cl^-, $S–SO_4^{2-}$, $N–NO_3^-$, $P–PO_4^{3-}$, $N–NH_4^+$, Ca^{2+}, Mg^{2+}, Na^+, K^+ and HCO_3^-, in precipitation and stream water were measured using ion chromatography, while suspended sediment during floods was measured using the 1 L bottle method. A significant increase in air temperature (+0.7°C per decade), especially during winter, led to a significant decrease in snowfall (SF) (−29.1 mm per decade) and snow cover duration (−16 days per decade), resulting in altered seasonal runoff patterns and an increase in the duration of hydrological drought. Decreases in ion concentrations and a 23% reduction in chemical denudation over the last decade highlight the effect of reduced anthropogenic pressures. Nutrient loads have also decreased by 38% due to improved wastewater management and agricultural abandonment, which has also resulted in a 60% reduction in suspended sediment loads. Mechanical denudation remains strongly linked to extreme hydrometeorological events.

Keywords: atmospheric deposition, chemical and mechanical denudation, suspended sediment, land use changes, ion chromatography, Polish Carpathians

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Introduction

The extent of chemical and mechanical denudation in catchments is crucial to understanding the intensity of the processes that shape and change these landscapes over time (Kostrzewski, Zwoliński 1992, Larsen et al. 2014, Cendrero et al. 2022). Denudation processes involve the removal of rock material from the Earth's surface through erosion, changes in relief and the transport of rock material. The effects of chemical and mechanical denudation can be observed locally, regionally and globally by calculating the flux of sediment and solute in the stream or river (Caine 2004, Milliman, Farnsworth 2013, Larsen et al. 2014, Cendrero et al. 2022, Kijowska-Strugała, Bochenek 2023). However, the total discharge of sediment and solutes does not represent all of the geomorphic work that occurs within a catchment (Caine 2004).

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Denudation plays a particularly important role, especially in mountainous areas (Montgomery, Brandon 2002, Caine 2004, Vergara et al. 2024), which can lead to a disproportionate source of sediment and dissolved matter transported to the seas and oceans (Milliman, Syvitski 1992, Syvitski et al. 2022). Studies by Larsen et al. (2014) have shown that >50% of total denudation and 40% of total chemical denudation occur in the steepest approximately 10% of the Earth's land surface. Chemical denudation is dependent on many environmental factors, including relief, geology, soils and hydrometeorological conditions. In many areas, it is strongly influenced by human activity, which can alter the direction and rate of change (Beylich, Laute 2004, Rivas et al. 2006, Walling 2006, Syvitski et al. 2020, 2022). One of the most important hydrometeorological factors that affect denudation is extreme events (Kostrzewski et al. 1992, Tomkins et al. 2007, Ciupa 2008, Gomes et al. 2022), including floods, which can cause river bed deepening (Lach, Wyżga 2002, Krzemień 2003), and bank erosion (Beylich, Laute 2021, Hamidifar et al. 2024). Anthropogenic changes in catchments, such as the development of infrastructure, deforestation or land use changes, can contribute to the intensification of mechanical erosion, the disruption of sediment flows and the reduction of natural water retention (Wemple et al. 2001, Syvitski et al. 2005, Prokop, Sarkar 2012, Zwoliński et al. 2018, Chen et al. 2019, Liu et al. 2021). Long-term studies in experimental plots in the Carpathians across different types of land use highlight the significant influence of land use on the denudation rate (Gil et al. 2021).

The connectivity and/or disconnectivity between slopes and channel systems have been the subject of intense debate. Studies by Święchowicz (2002) and Phillips (2003) show that land use change in mountainous catchments has no direct effect on sediment movement within the said catchment. Richards (2002) also highlighted the important influence of catchment buffers (e.g. floodplains) on the amount of denudation. Studies by Oygarden et al. (2003), Latocha (2006) and Bucała, Budek (2011) have shown that the reduction of cultivated land and the increase of forested areas and stream channel regulation can intensify channel deepening. Slope and channel systems are often linked by roads, which have the highest runoff coefficient (Rc) of all land use types (Soja 2002). The presence of unpaved roads in a catchment can increase sediment loads by up to 80% (Froehlich 1986). Switalski et al. (2004) and Keppeler and Lewis (2007) noted that roads can continue generating sediment for many years after their construction. Roads also affect the chemical composition of water (Kelly et al. 2008, Corsi et al. 2010), are a source of heavy metals (Dall'Osto et al. 2014) and increase water salinity due to the use of de-icing salts (Szklarek et al. 2022, Płaczkowska et al. 2024).

Despite numerous studies conducted across different areas worldwide, the issue of denudation is still relevant due to ongoing climate change, including an increase in the frequency and severity of extreme events as well as various forms of anthropopressure. Understanding the interaction between natural and anthropogenic factors in mountainous regions allows for the more accurate prediction of environmental changes as well as the formulation of conservation strategies based on long-term measurement data. This study aims towards the following:

1. Determine the magnitude and dynamics of water discharge and transported substances (including nutrients and suspended sediments) in a small mountain catchment between 1995 and 2023;
2. Identify the main factors influencing fluvial transport variability as well as the regularities that shape long-term trends, changes and relations.

Materials and methods

Study area

This study was carried out in the Bystrzanka catchment (49°37′51″N, 21°07′02″E) located in the Polish Flysch Carpathians (Fig. 1A, Starkel 1972). The study area has an average altitude of 487 m a.s.l. and an average slope of 9.6°. Slopes reach up to 35° in the western part of the catchment, primarily facing NE, while slopes in the eastern region only reach up to 15° and primarily face SW (Fig. 1B). The geology of the studied area is dominated by rocks from the Magura Nappe (Kozikowski 1956, Świdziński 1973). The occurrence of differentiated lithological complexes in the catchment is of particular importance. The western section of the catchment (600–753 m a.s.l.) is characterised by the occurrence of coarse- to medium-bedded sandstone and shale formations from the Magura series, which are characterised by their high resistance to denudation processes. The eastern section of the basin is composed of less resistant shale–sandstone inoceram layers (thin-, medium- and thick-bedded sandstones with shale and fucoid marls) from the Cretaceous–Paleogene period (Kopciowski et al. 1997, Fig. 1C). The presence of shale in the geological structure as well as the occurrence of weathered cover on the slopes favour denudation processes (Kozikowski 1956, Świdziński 1973). The bedrock in the catchment has undergone secondary folding and has been displaced along the Ropa axis fault – it is also cut by minor faults (Świdziński 1953, Sikora 1970). Around 30% of the catchment area consists of landslides.

The dominant soil types in the catchment are sandy clay loam and clay loam Inceptisols with a relatively low water capacity (Soil Survey Division Staff 2017). The thickness of the soil cover is 80–100 cm near the hilltops and increases to 200–400 cm along the slopes. Soils have formed on the slope cover, which – in areas built on Magurian sandstones – have a stony fraction of approximately 50–80%. In contrast, in the shale–sandstone series of the Inoceramian and Krosno beds, the proportion of skeletal fractions is 20–40% (Adamczyk et al. 1973).

The main course of the Bystrzanka valley has a narrow bottom (rocky-alluvial) and steep slopes of up to 15 m; the valley is about 3–4 m wide in its middle course, and the riverbed is about 10 m wide at the mouth of the Bystrzanka valley where it leads into the Ropa (Niemirowska 1970, Kijowska-Strugała 2015).

Over time, the catchment has undergone significant changes in land use and land cover (LULC), primarily due to a major political transformation in 1989, which had a significant effect on southeast Poland. The eastern part of the catchment area has been used for agricultural purposes for many years. In 1969, cultivated land accounted for 48% of the area, while meadows made up only 3%. However, fundamental changes in the LULC have been observed in the past 60 years, with the area of cultivated land decreasing by up to 80% with a complementary eightfold increase

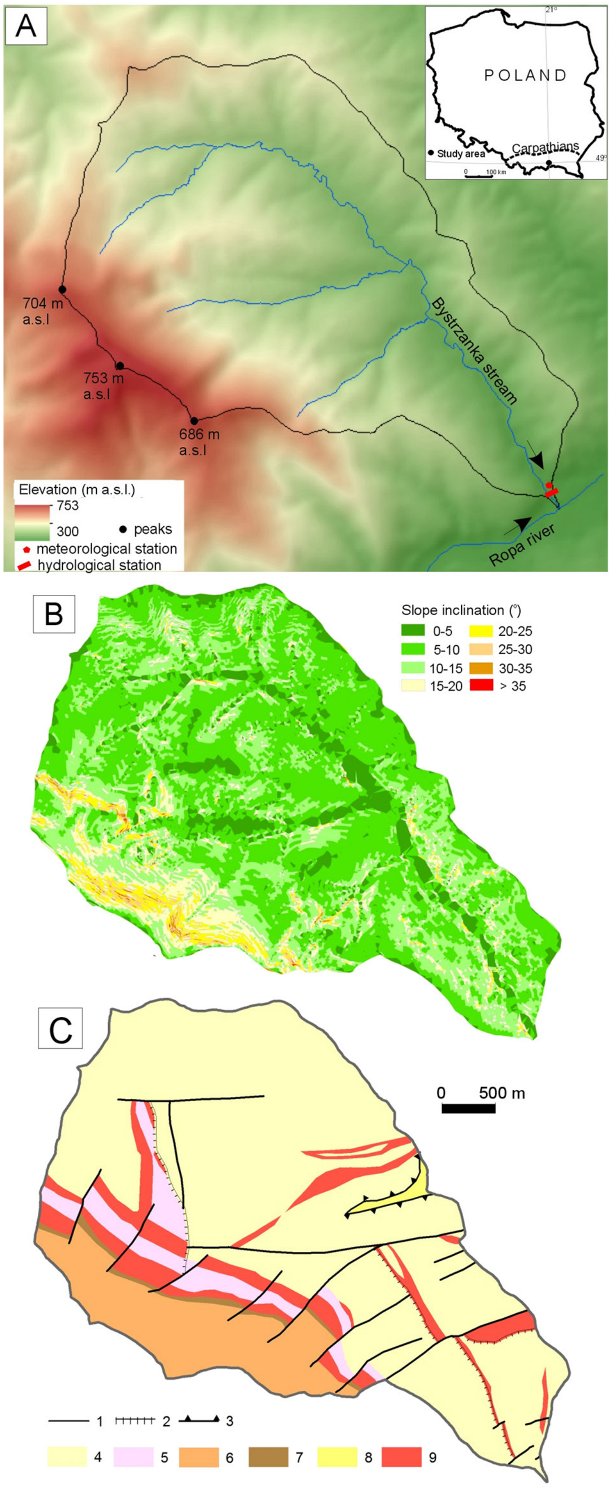

Fig. 1. A – Location of the study area and environmental characteristics of the Bystrzanka catchment area; B – slope; based on the DEM with 10 m resolution; C – geological structure: 1– fault, 2 – minor thrust fault, 3 – major thrust fault, 4 – Inoceramus beds, 5 – Ciężkowickie sandstones, 6 – Magura sandstones, 7 – Sub-Magura sandstones, 8 – Krosno Bed, 9 – variegated shales; based on Wójcik et al. (2003).

in meadow area (Fig. 2). European Union (EU) policy supports the mowing of meadows, for example, for use as fodder. Two villages within the catchment area, Szymbark and Bystra, have a combined population of around 5000 people (Central Statistical Office of Poland 2022). The topographic database (BDOT10k – Polish acronym) of the catchment area indicates that the 448 buildings within the region are primarily concentrated along the valley axis. Over the past 50 years, an increase in the population and development of the two villages have led to a 19% rise in road density (Kijowska-Strugała 2019), highlighting how LULC and population density have contributed to changes in the area and spatial distribution of road surfaces in the region. The construction of new paved roads in the valley bottom has been accompanied by the abandonment of agricultural fields and cart roads on slopes. In the early 21st century, particularly in 2009 and 2010, the catchment saw a surge in engineering investments co-financed by EU funds. During this time, ditches along the main road were sealed, six landslides were stabilized, the largest bridge over the main stream was reconstructed and a 2-km section near the mouth of the Bystrzanka stream was reinforced with concrete (Kijowska-Strugała 2019, Gorlice Municipal Office 2024).

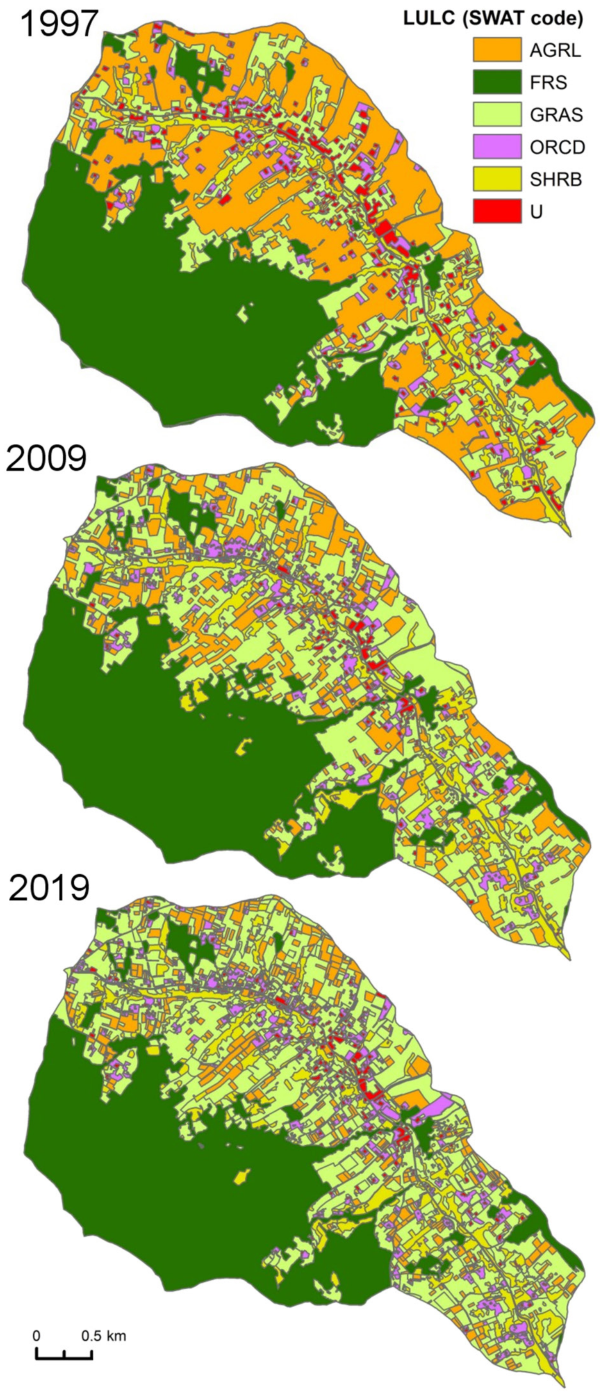

Fig. 2. Land use an land cover (LULC) changes in Bystrzanka catchment (1 – cultivated land, 2 – forest (deciduous, mixed), 3 – grassland, 4 – orchards, 5 – agro-forestry transitional areas, 6 – urbanised area, industrial or commercial, sports or recreational areas, low-density buildings, roads; based on aerial photos Olędzki 2007, Zwoliński, Gudowicz 2011, Bochenek 2020).

Methods

Studies of meteorological and hydrological parameters have been carried out continuously from the meteorological station that is part of the Szymbark Research Station (SRS) of the Institute of Geography and Spatial Organization of the Polish Academy of Sciences since the 1970s. The SRS is located in the Bystrzanka catchment. The meteorological station is located at an altitude of 325 m a.s.l. This study used data from 1995 to 2023. Daily precipitation totals (wet precipitation and dry deposition) were determined using a Hellman rain gauge and an automatic VAISALA station. Due to the lack of other precipitation stations in the study area, the results of a single station located near the mouth of the Bystrzanka stream into the Ropa river were taken into account; this is despite the likelihood of a precipitation gradient in the study area as well as increased precipitation with altitude. Bochenek (2007) analysed the spatial differentiation between the total annual and seasonal precipitation in the Beskid Niski, Ciężkowice Foothills and Jasielsko-Sanockie Basin between 1970 and

1981 based on data from 27 precipitation stations. The calculated gradient of annual precipitation was 30 mm per 100 m of altitude. Based on this profile, precipitation at the highest point of the catchment (753 m a.s.l.) should be about 130 mm higher than the meteorological station, averaging around 995 mm.

The Olechnowicz-Bobrowska et al. (1970) classification was used to assess the frequency of days with a certain amount of precipitation. The ionic composition of precipitation water was determined from monthly samples of total deposition, derived from the daily precipitation totals.

Water level changes in the stream were recorded using a KB2 limnigraph and converted into flow rates. The flow curve was updated after each flood, especially when changes in bed morphology were observed. The Rc was calculated as the quotient of runoff (annual values) and precipitation (annual values). Floods and hydrological droughts were identified based on the magnitude of the discharge. The criterion proposed by Ozga-Zielińska and Brzeziński (1997) was used to identify flood events. Floods were also grouped according to their origin based on the terminology described by Lambor (1965). The lower discharge threshold for flood events was set at 0.689 $m^3 \cdot s^{-1}$. Hydrological droughts were identified using the method proposed by Hisdal et al. (2004), with the threshold set at the 0.70 quantile. Over the study period, this upper threshold was 0.035 $m^3 \cdot s^{-1}$.

Chemical and mechanical denudation studies have been carried out in the Bystrzanka catchment since the 1970s and continuously since 1995. Water temperature, pH and electrical conductivity (EC) relative to 25°C were measured daily using an Elmetron (CP-315, CC-311 and CX701) over the entire study period. Both pH and EC were calculated as monthly weighted averages with respect to precipitation or water flow in the channel. Water samples were collected at the mouth of the stream for chemical analysis. During floods, the frequency of water sampling was adjusted based on the rate of change in water level in the channel (from 15 min to about 1 h). The water samples were filtered using Whatman glass microfibre filters (GF/D) 2.7 µm and their ionic composition was determined using ion chromatography (Dionex ICS 1100) (Cl^-, S–SO_4^{2-}, N–NO_3^-, P–PO_4^{3-}, N–NH_4^+) and atomic absorption spectrophotometry (VARIAN) (Ca^{2+}, Mg^{2+}, Na^+, K^+). All ions have been identified using ion chromatography since 2015. As other studies have shown, both spectrophotometric and ion chromatographic methods give very similar results (Thienpont et al. 1996). The concentration of HCO_3^- in the stream was measured by titration with 0.1M HCl. The chemical composition in the runoff (R) and precipitation (P) was calculated by multiplying the weighted ion concentration by the runoff or precipitation, respectively. The dissolved load (DL; $kg \cdot a^{-1}$) and yield (DY; $kg \cdot ha^{-1} \cdot a^{-1}$) in the precipitation (DL_P; DY_P) and runoff (DL_R; DY_R) were also determined. The chemical denudation balance ($kg \cdot ha^{-1}$) was calculated as the difference between DL_R and atmospheric input (DL_P).

Suspended sediment concentration (SSC) was measured manually using the 1 L bottle method (Wren et al. 2000) in the hydrometric profile at 60% water depth as recommended by Newburn (1988). The frequency of sampling was approximately every 15–20 min during floods and decreased with decreasing water levels. During periods of low-intensity rainfall, which caused the water level in the stream to rise slowly, samples were taken every 1–2 h. Sampling frequencies were relatively consistent during the study period. Daily water samples were collected at 6 am (UTC) as part of the continuous monitoring of the stream. The presence of SSC was not observed during periods of average and low water levels in the Bystrzanka stream.

The water samples were filtered; any sediment accumulated on the filters was dried (105°C) and weighed to determine the SSC ($g \cdot dm^{-3}$) in the laboratory at the SRS. The area-specific suspended sediment yield (SSY; $Mg \cdot km^{-2} \cdot a^{-1}$) and absolute suspended sediment load (SSL; $Mg \cdot a^{-1}$) were calculated based on the SSC using the following equation:

$$SSL = \sum_1^n SSC \cdot Q \cdot \Delta t \qquad (1)$$

where:
- n – number of samples;
- SSC – suspended sediment concentration ($kg \cdot m^{-3}$);
- Q – average discharge ($m^3 \cdot s^{-1}$);
- Δt – time interval.

Statistical analyses were performed using Statistica v.12. The Shapiro–Wilk distribution test was used to assess the distribution of data. The non-parametric Mann–Whitney *U* test was used to compare the differences between the first (1995–2004) and last (2014–2023) decades of the research period. The Pettitt test was used to detect changes in the chemical denudation balance data. This method identifies a single point of change in the time series, that is, the point at which there is a significant change in the trend or mean of the distribution of the variable. The data were statistically analysed using a linear trend. A significance level of $a = 0.05$ was applied for trend analysis, as is standard in statistical research (e.g. Souvignet et al. 2012). For results with higher statistical significance and more pronounced trends, values were reported with a higher significance level (i.e. $p < 0.01$).

Results

Hydrometeorological conditions

The mean annual air temperature during the study period was 8.8°C and exhibited a statistically significant increasing trend of 0.7°C per decade (α = 0.05; Table 1). A positive trend in mean monthly air temperature was observed in all months of the year except May (Table 1).

The mean annual precipitation during the study period was 864.5 mm, with annual totals ranging from 612.4 mm (2003) to 1180.0 mm (2014) (Table 1 and Fig. 3). The mean monthly precipitation ranged from 42.0 mm in December to 130.6 mm in July. Precipitation totals fluctuated each month between 1995 and 2022. The largest increase in precipitation occurred in August (12.5 mm per decade), while the largest decrease occurred in June (−18.1 mm per decade). The trend coefficient was influenced by the monthly precipitation totals and both annual and monthly rainfall totals were not statistically significant and did not show directions of change over the multi-year period. The average number of days with precipitation (Np ≥0.1 mm) was 178 days · a^{-1}. In this context, a significant change was observed in 2012. Between 1995 and 2012, the average number of days with precipitation was 184 days · a^{-1} and exhibited a statistically significant positive trend of 12 days per decade. However, after 2012, the number of days with precipitation declined significantly, with a negative trend of 72 days per decade. This sharp decrease only stopped in 2020. The average number of days with precipitation between 2012 and 2023 was 170 days · a^{-1}.

The structure of daily precipitation was dominated by very light (≤1.0 mm) and light (1.0 < ... 1.0 < P ≤ 5.0) precipitation, accounting for 32.2% and 41.5% of the days, respectively. The decrease in the number of days with precipitation between 2014 and 2023 was accompanied by

Table 1. Statistical characteristics of temperature (T), precipitation (P), snowfall (SF), runoff (R) and Rc in the Bystrzanka catchment between 1995 and 2023 (bold: statistically significant $p < 0.05$).

Period	Air temperature T		Precipitation P		Snowfall SF		Runoff R		Runoff coefficient Rc
	Average	Trend (per decade)	Average	Trend (per decade)	Average	Trend (per decade)	Average	Trend (per decade)	
	[°C]		[mm]						[%]
Year (1995–2023) (min–max)	8.8 (6.2–10.1)	**0.73**	864.5 (612.4–1180.0)	−4.5	118.2 (17.0–222.8)	**−25.0**	390.2 (85.0–856.6)	**−74.5**	45 (13–73)
Cv (%)	10.5		15.5		42.3		35.2		
Winter half-year (1995–2023)	2.3	**0.91**	283.2	−8.2	118.2 (17.0–222.8)	**−25.0**	199.3	**−43.5**	70
Summer half-year (1995–2023)	15.2	**0.51**	581.3	3.7			190.9	−31.0	33
Year (1995–2004)	8.0		850.2		138.3		441.4		52
Year (2014–2023)	9.6		880.0		77.3		317.7		36

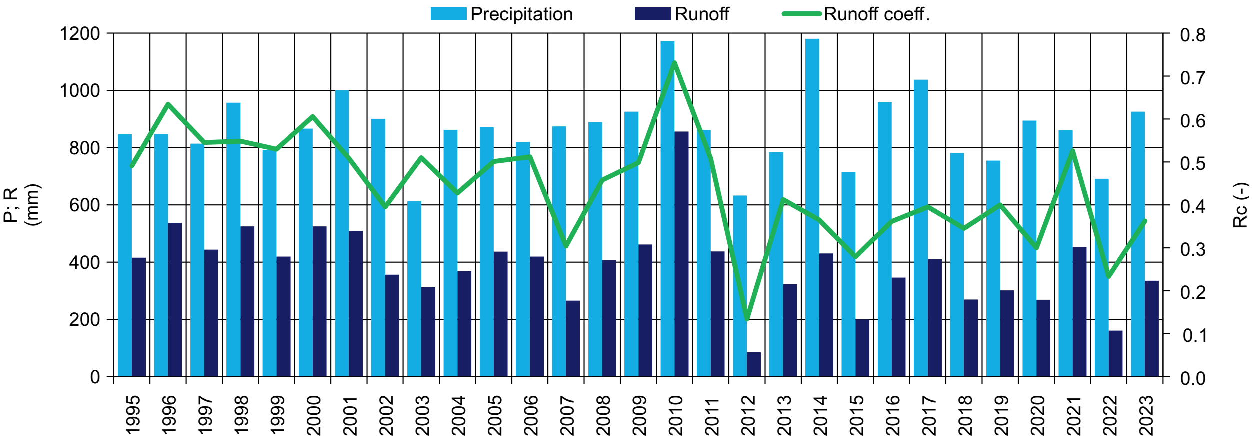

Fig. 3. Annual totals of precipitation (P), runoff totals (R) and runoff coefficient (Rc) in 1995–2023.

a decrease in the number of days with very light precipitation. Between 1995 and 2022, there was an increase in the number of days with precipitation between 20 mm and 30 mm. Such precipitation, especially over periods of several days, resulted in significantly increased streamflow.

Differences in daily precipitation, annual snowfall (SF) and snowpack accumulation patterns observed in the first and last decades of the study have been attributed to climate change. The increase in air temperature, particularly during winter, has contributed to a negative trend in annual SF totals, despite the lack of any significant changes in precipitation during this hydrological half-year (HY). This warming has also led to a decrease in snow cover as well as the shortening of the snow cover retention period (Table 2).

This significant increase in temperature during the winter months has also affected the amount of SF and the duration of snow cover. The average annual SF was 117.6 mm (13.6% of total precipitation), while the average duration of snow cover was 67 days · a^{-1}. The highest SF

Table 2. Number of days with precipitation and snow cover in 1995–2023.

Precipitation (P)	1995–2023	1995–2004	2014–2023
	Number of days		
Total	181	184	165
≤1.0 mm	66	68	52
1.0 < P ≤ 5.0 mm	64	67	61
5.0 < P ≤ 10.0 mm	26	26	26
10.0 < P ≤ 20.0 mm	17	16	17
20.0 < P ≤ 30.0 mm	5	3	6
30.0 < P ≤ 50.0 mm	3	3	3
>50.0 mm	1	1	1
Snow cover	67	81	45

totals were recorded in the winters of 1999/2000 (222.8 mm) and 2012/2013 (214.0 mm), while the lowest totals were recorded in the winters of 2013/2014 (26.0 mm) and 2019/2020 (17.0 mm) (Fig. 4). Over the study period, there was a statistically significant negative trend in both annual SF (−29.1 mm per decade) and snow cover length (−16 days per decade).

The mean annual runoff from the Bystrzanka catchment was 390.2 mm, representing 45.1% of the total precipitation (Table 1). The total annual runoff ranged between 85.0 mm in 2012 and 856.6 mm in 2010 (Fig. 3), with a coefficient of variation (Cv) of 36.8%. The mean runoff in the winter hydrological HY was 199.3 mm (51.1% of the annual total; Cv = 36.6%), while the mean runoff in the summer hydrological HY was 190.9 mm (48.9% of the annual total; Cv = 56.6%). The proportion of runoff in the winter hydrological HY compared with the annual total ranged from 22.8% in 2014 to 75.2% in 2022.

A total of 294 floods occurred in the main stream between 1995 and 2023. Floods resulting from continuous rainfall and from downpours were relatively similar – 95 and 94 events, respectively (Fig. 5A). The total flood discharge (i.e. the amount of water above the threshold discharge) was 2575 mm, which accounted for 22.8% of the total discharge. Floods after downpours discharged the most water (998.8 mm), while mixed floods (i.e. due to a combination of rainfall and snowmelt) contributed the least discard (229.2 mm; Fig. 5B). The mean annual flood runoff was 89.4 mm. The number of floods per year ranged from 2 (2012) to 22 (2010), while the magnitude of the flood runoff (water level above

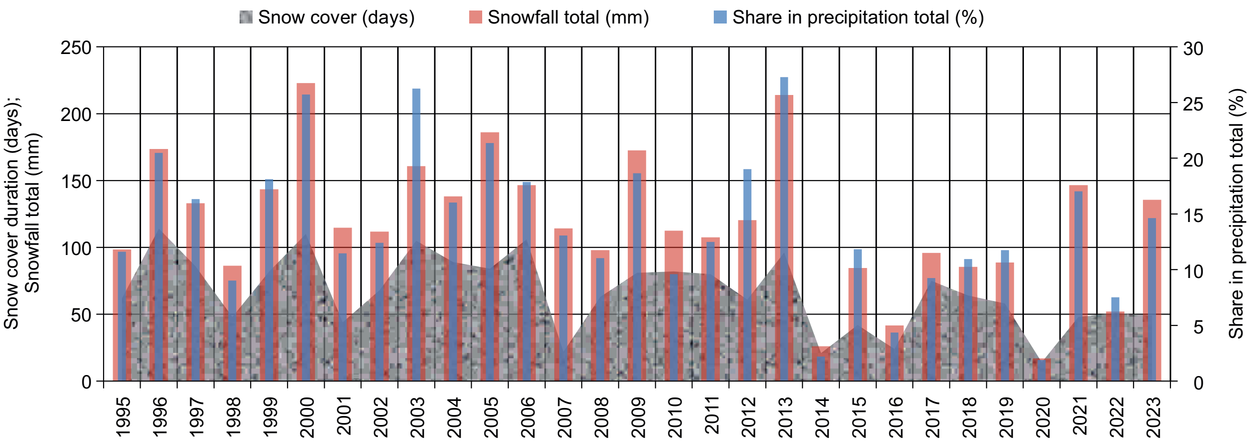

Fig. 4. Annual SF, share of snowfall in annual precipitation and duration of snow cover. SF, snowfall.

discharge threshold) ranged from 3.2 mm (2022) to 263.5 mm (2010) (Fig. 6). Between 2014 and 2023, there was an average of 9.7 floods, 1 less than between 1995 and 2004, and flood runoff was 29 mm lower. The duration of hydrological droughts ranged from 64 days (1998) to 303 days (2012). On average, hydrological droughts occurred on around 163 days · a^{-1}, including 64 deep drought days · a^{-1}. Over the period analysed, a statistically significant increase in the duration of hydrological droughts was observed (p = 0.033). In 1995–2004, they lasted 136 days on average, and 42 days longer in the last decade.

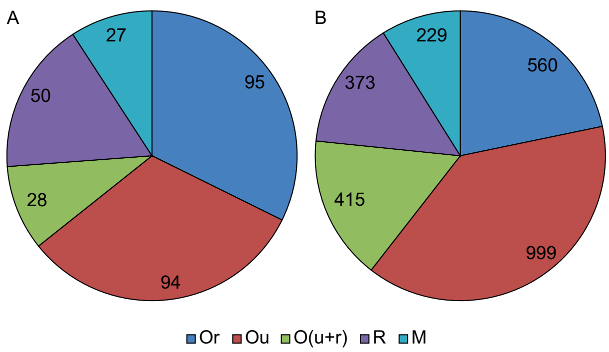

Fig. 5. A – Number of floods distinguished by genetic classification (Lambor 1965) in 1995–2023; B – layer of water above discharge threshold (mm). Or – continuous rainfall, Ou – heavy rainfall, O(u + r) – mixed rainfall, R – snowmelt, M – mixed rainfall and snowmelt.

Temporal variation of chemical and mechanical denudation

Chemical composition of precipitation

The mean annual pH of precipitation exhibited a statistically significant increasing trend of 0.3 units per 10 years (α = 0.05), with values ranging from 3.99 (2003) to 5.60 (2022) (Fig. 7; mean annual value = 4.90; Cv = 8.2%). However, there was no statistically significant trend observed in the mean annual EC (multi-year mean = 1.90 mS · m^{-1}; range of variation 0.90–5.60 mS · m^{-1}).

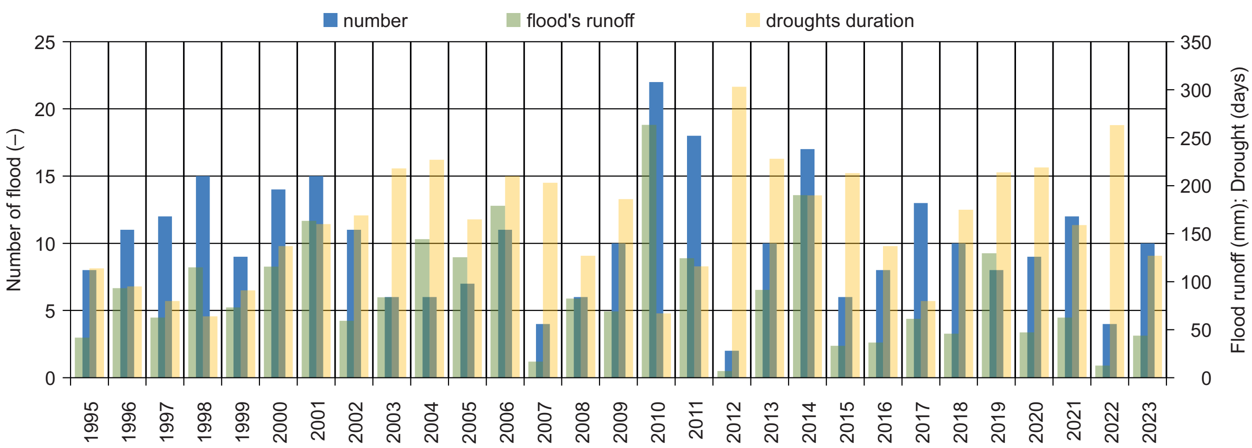

Fig. 6. Flood runoff and number and hydrological drought duration from 1995 to 2023.

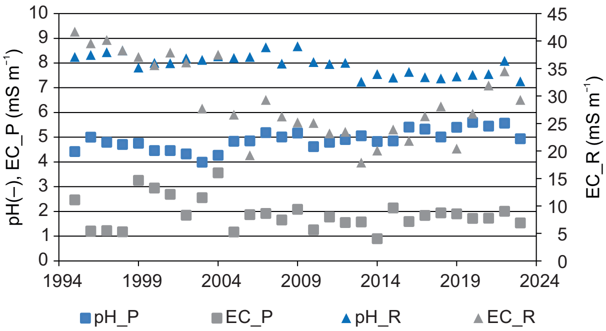

Fig. 7. Mean annual pH and electrical conductivity (EC) in precipitation (P) and runoff (R).

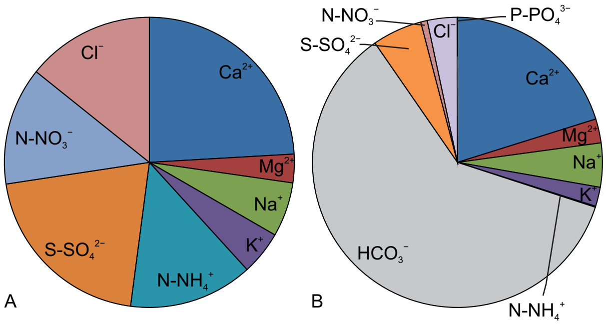

Fig. 8. Average ion concentration (%) of in A – precipitation and B – runoff.

The ionic composition of the precipitation was dominated by Ca^{2+} and $S–SO_4^{2-}$ (Fig. 8A). The mean DL_P over the multi-year period was 42.39 kg · ha^{-1} (ranging between 29.2 kg · ha^{-1} and 83.7 kg · ha^{-1}). The lowest DY_P recorded was for Mg^{2+} (1.61 kg · ha^{-1} · a^{-1}), while the highest was for Ca^{2+} (10.16 kg · ha^{-1}) (Fig. 9); this is related to the 'washing out' of the troposphere by precipitation after increasingly long periods without precipitation due to the accumulation of fine dust in the troposphere (Table 3). $N–NO_3^-$ exhibited the lowest coefficient of variation in mean annual values (32%), while the highest value was associated with Cl^- (87.5%). There were statistically significant differences ($p < 0.05$) in Na^+, K^+, $N–NH_4^+$ and pH between the first and the last decades of the study period. In addition, higher mean monthly concentrations of Cl^-, Mg^{2+}, Na^+, $N–NO_3^-$ and $S–SO_4^{2-}$ were observed in winter compared with the warmer periods.

Chemical composition of the runoff

The mean annual pH in the runoff was 7.94 (ranging between 7.24 and 8.68; *Cv* = 5.3%) and

Table 3. Characteristics of dissolved ion in dissolved yield precipitation (DY_P) and dissolved yield runoff water (DY_R) from 1995 to 2023 (bold: statistically significant $p < 0.05$).

Parameter	Precipitation			Runoff		
	DY_P [kg · ha^{-1} · a^{-1}]	Cv [%]	Coefficient	DY_R [kg · ha^{-1} · a^{-1}]	Cv [%]	Coefficient
$S–SO_4^{2-}$	8.62	40.3	**−0.18**	48.38	45.7	**−1.06**
$N–NO_3^-$	5.50	32.0	−0.05	5.90	39.8	**−0.18**
Cl^-	6.61	87.5	−0.05	28.48	40.2	−0.39
Na^+	2.54	63.9	**0.09**	18.60	33.7	−0.19
K^+	2.03	37.2	0.03	18.60	40.0	**−0.42**
Mg^{2+}	1.33	45.8	0.01	23.33	40.4	**−0.50**
Ca^{2+}	10.16	39.8	**0.21**	175.24	38.9	−2.54
$N–NH_4^+$	5.60	37.3	**−0.19**	0.89	92.7	**−0.05**
HCO_3^-				516.56	33.4	−4.83
$P–PO_4^{3-}$				0.41	66.5	**−0.02**
Load (1995–2023)	4.90	28.6	−0.12	859.9	33.0	**−0.14**
1995–2004 vs. 2014–2023						
p-value (Mann–Whitney U test)						
$S–SO_4^{2-}$	0.054			**0.004**		
$N–NO_3^-$	0.241			**0.000**		
Cl^-	0.076			**0.031**		
Na^+	**0.002**			0.273		
K^+	**0.045**			**0.002**		
Mg^{2+}	0.307			**0.017**		
Ca^{2+}	0.850			**0.021**		
$N–NH_4^+$	3.062			**0.017**		
HCO_3^-				0.162		
$P–PO_4^{3-}$				**0.000**		

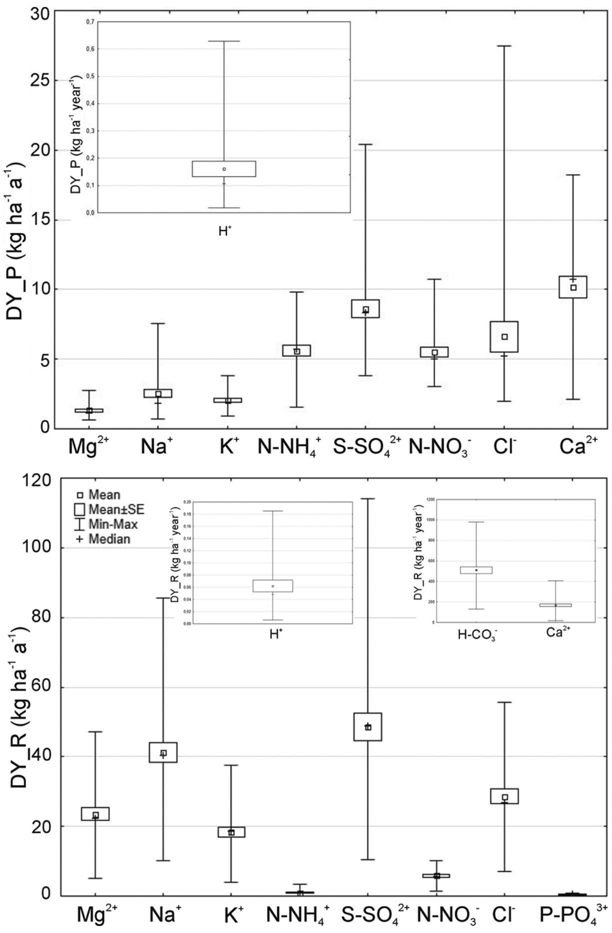

Fig. 9. Variation in the content of individual ions in dissolved yield precipitation (DY_P) and dissolved yield runoff (DY_R) between 1995 and 2023.

exhibited a statistically significant decreasing trend of 0.3 per 10 years (α = 0.05). Similarly, the mean annual EC was 29.8 mS · m^{-1} (ranging from 17.5 mS · m^{-1} to 41.7 mS · m^{-1}; Cv = 25.2%), with a decreasing trend of 5.1 mS · m^{-1} per decade. The mean annual DL_R was 859.9 kg · ha^{-1}, ranging from 206.1 kg · ha^{-1} (2012) to 1729.3 kg · ha^{-1} (2010). The range of changes in the DL_R of individual ions over the multi-year study period is presented in Fig. 8B. The dominant ion in the Bystrzanka water was HCO_3^- (Fig. 9; average of 516.56 kg · ha^{-1} · a^{-1}; Cv = 33.4%), though the ionic load of this species decreased by 17% in the last decade (2014–2023) compared with the first decade (1995–2004). A decreasing trend in the DY_R was observed for all ions and, in the case of S–SO_4^{2-}, N–NO_3^-, K^+, Mg^{2+}, N–NH_4^+, and P–PO_4^{3-} (Table 3), it was statistically significant. In addition, statistically significant differences ($p < 0.01$) were found in the DY_R between the first and last decades of the study period (except for Na^+ and HCO_3^-). There was a statistically significant decrease in the mean monthly load of all species due to the decrease in water flow (except for N–NO_3^-, N–NH_4^+). Furthermore, the loads of Cl^-, N–NO_3^- and S–SO_4^{2-} were higher in the colder half of the year compared with the warmer half of the year by 10%, 65% and 11%, respectively. This was influenced by the runoff regime (slightly higher but more stable runoff in winter) as well as the input of organic mineralisation products from the catchment.

The mean value of the total nutrient concentration (N–NH_4^+, N–NO_3 and P–PO_4^{3-}) for the studied period in the Bystrzanka stream was 1.94 mg · dm^{-3}. There was a statistically significant

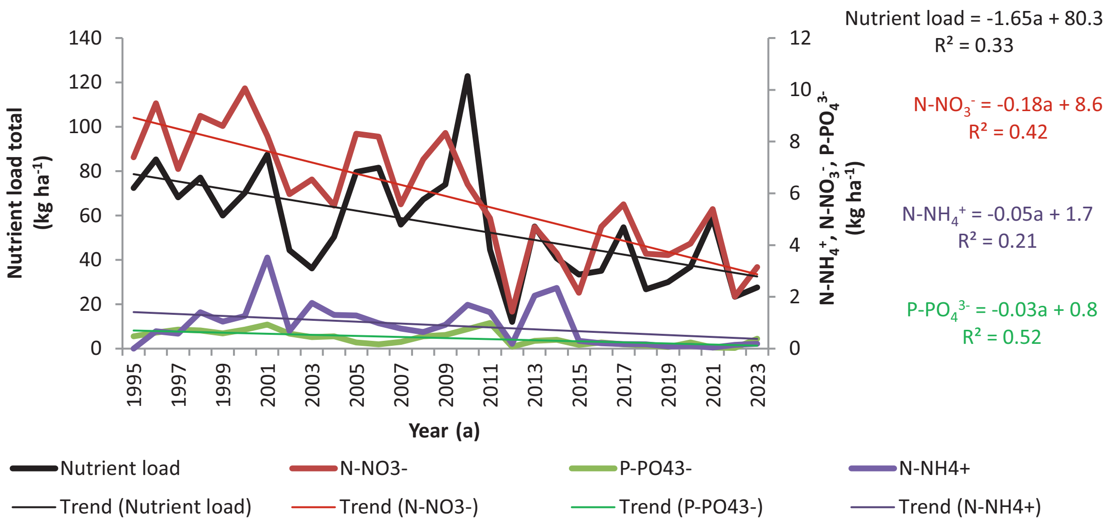

Fig. 10. Changes in individual nutrients (P–PO_4^{3-}, N–NO_3^-, N–NH_4^+) and total nutrient loads in the Bystrzanka stream between 1995 and 2023.

decrease in nutrient loads during the study period (Fig. 10). The average nutrient load in the first decade was 2.22 mg · dm^{-1}, which decreased by 38% in the period between 2014 and 2023.

Chemical denudation balance in the Bystrzanka catchment

The chemical denudation balance varied across the multi-year study period (Fig. 11), ranging from 177 kg · ha^{-1} (2012) to 1688 kg · ha^{-1} (2010) with a mean of 817 kg · ha^{-1}. The year 2012 was an extremely dry year with a Rc of only 13.4%, while 2010 was characterised by record runoff (Rc = 73.1%). Statistical analyses (Pettitt test) revealed a statistically significant point of change in the chemical denudation balance in 2011. The average denudation value before 2011 was 939 kg · ha^{-1}; in the years following, this value decreased to 668 kg · ha^{-1}. The reason for the changes may have been lower precipitation since 2011 and lower average runoff by 32%. In addition, changes in LULC affected the amount of dissolved matter carried out from the catchment. A comparison of the two periods (i.e. the first and last decades) exhibited statistically significant differences (p = 0.045). The average value

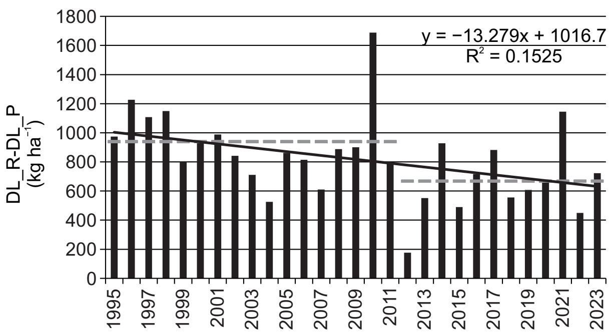

Fig. 11. Chemical denudation balance in the Bystrzanka catchment between 1995 and 2023.

in the first decade was 927 kg · ha^{-1}, 23% higher than in 2014–2023.

A multi-year decrease in DL_R was observed for all ions, while there were multidirectional changes observed in the DL_P: Na^+, K^+, Ca^{2+} and Mg^{2+} loads increased, while N–NO_3^-, S–SO_4^{2-} and N–NH_4^+ loads decreased. The decrease in the chemical denudation balance was primarily influenced by changes in the ion composition of the stream.

Suspended sediment in the stream

SSC values varied across multiple flood events, reaching values of up to 45 g · dm^{-3} during the extreme flood in 2010, when flows reached 57.7 m^3 · s^{-1} (Fig. 12A). Such high SSC values were associated with natural factors (high daily precipitation and record flow) combined with anthropogenic factors, including engineering works that resulted in the accumulation of loose material both near and within the riverbed itself. Higher SSC values were recorded during summer floods over the study period. In addition, the SSC peaked both before (clockwise loop – supply of material mainly from the channel; Fig. 12A) and after (counter-clockwise loop) the peak discharge (Fig. 12B). Higher SSC values following peak discharge are a result of the delayed delivery of material from the upper part of the catchment, primarily as a product of the erosion of the stream channel and its tributaries, the erosion of roads and roadside ditches and from material accumulated in the vicinity of the channel due to anthropogenic activities.

The mean SSL between 1995 and 2023 was 3521 Mg · a^{-1} (ranging from 48 Mg · a^{-1} to 21,658 Mg · a^{-1}) (Fig. 13). Statistical analysis (Pettitt test) revealed a statistically significant point of

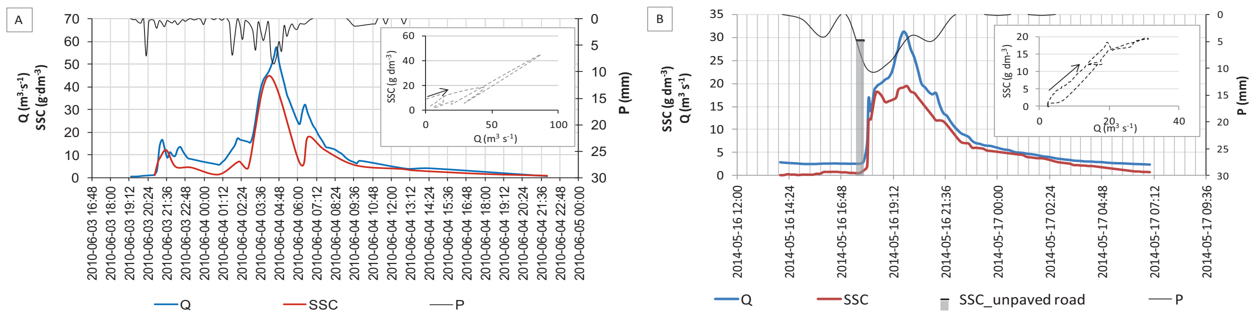

Fig. 12. Plot of SSC in the stream against discharge (Q) and precipitation (P) during the A – 3–4 June 2010 and B – 16–17 May 2014 flood events with a hysteresis loop and an example of concentration on an unpaved road in the Bystrzanka catchment. SSC, suspended sediment concentration.

change in the SSL regime in 2016. The year 2010 experienced the highest SSY of 1666 Mg km^{-2} · a^{-1} and the highest stream runoff of 896.7 mm: 95.8% of the annual SSL was removed during two consecutive flood events (May–June).

There was a statistically significant relationship between the runoff above the flood threshold and the SSY during the analysed period (Fig. 14). In addition, the SSY was found to be 317 Mg · km^{-2} · a^{-1} in the first decade (1995–2004); this was halved in the last decade of the study period (2014–2023; 125 Mg · km^{-2} · a^{-1}).

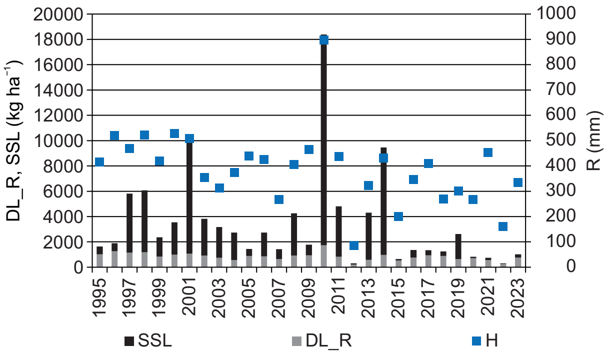

Fig. 13. DL_R and SSL changes against runoff (R) between 1995 and 2023. DL_R, dissolved load runoff.

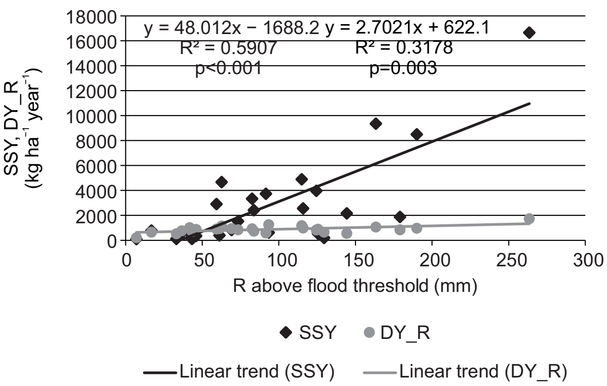

Fig. 14. Dependence of SSY and DY_R on the runoff above the flood threshold between 1995 and 2023. DY_R, dissolved yield runoff.

Discussion

Mountains are the areas with the highest denudation rates (Larsen et al. 2014); the rate of denudation in the Polish Carpathians is as high as 90 mm per 1000 years (Pulina 1974). In addition to environmental factors, the rate of denudation is significantly affected by climate change (Mariotti et al. 2021, Cendrero et al. 2022). The integrated analysis of thermal, precipitation and hydrological data from the Bystrzanka catchment between 1995 and 2023 reveals a comprehensive pattern of climate-induced changes. A statistically significant increase in mean annual air temperature (+0.7°C per decade), which is particularly marked in the winter season, has led to a notable decline in annual SF totals and snow cover duration.

Increases in air temperature affect the intensity of convection, leading to an increase in the frequency of precipitation with higher daily totals and greater short-term intensity. Such patterns are commonly attributed to climate change and have led to an increase in catastrophic events, such as floods and hydrological droughts (Pińskwar et al. 2019, Szwed 2019). Despite the absence of a statistically significant long-term trend in the total precipitation within the Bystrzanka catchment, the number of days with precipitation ≤1 mm has decreased, while the frequency of intense rainfall events (20–30 mm · day^{-1}) has increased. These shifts have contributed to a reduction in the number of days with precipitation since 2012 as well as to a more irregular precipitation regime. The observed increase in high-intensity rainfall events, combined with the reduced snow retention, has influenced both flood frequency (294 events in total) as well as the intensity of flood runoff, particularly in years characterised by extreme precipitation (e.g. 2010). Conversely, years with reduced precipitation totals and higher temperatures (e.g. 2012) coincided with prolonged hydrological droughts. The interaction between rising air temperatures, altered precipitation patterns and hydrological responses highlights the vulnerability of small mountain catchments to climate change and emphasises the need for multi-parameter monitoring in water resource and risk management strategies.

There was a statistically significant increase in the pH of precipitation in the Bystrzanka catchment between 1995 and 2023. According to Bochenek and Szydłowski (2024), the average pH was found to be lower during anticyclonic atmospheric circulation, especially in winter, when water acidification increases due to pollutants from the Silesian conurbation, the Kraków agglomeration and nitrogen plants in Tarnów, as well as emissions from Slovakia. Ion deposition (DY_P) in the studied catchment was half of those observed in the Carpathian foothills (Żelazny 2005), where the impact of human activity is greater. The chemical denudation balance in the Bystrzanka catchment exhibited a significant downward trend, with a regression coefficient of 13.3 kg $\cdot$ ha^{-1} $\cdot$ a^{-1}.

Research by Cendrero et al. (2022) shows that human activity plays a dominant role in denudation rates and sediment yields. In the studied catchment, the collapse of the centrally planned economy and the introduction of a free market economy led to significant changes in LULC (Kijowska-Strugała, Bochenek 2023). The proportion of cultivated land decreased from 48% (1969) to 8% (2019), with the largest reduction occurring at the turn of the 20th and 21st centuries. Consequently, the average soil erosion on slopes was reduced by 73% based on the RUSLE model (Kijowska-Strugała 2019). Similar changes in LULC were observed in other countries belonging to the communist regime in Eastern Europe (Bezak, Mitchley 2014, Munteanu et al. 2014, Bucała-Hrabia 2017).

Since 1995, there has been a systematic decrease in the annual mean concentrations of dissolved nutrients in the stream (DY_R), indicating an improvement in the sanitary quality of the water; these trends are statistically significant ($p < 0.05$). The most important factors influencing these changes are the reduction of livestock, a decline in agricultural land and the use of fertilisers, and the modernisation of the infrastructure of domestic sewage systems in the lower part of the catchment (Gorlice Municipal Office 2024). Czekaj and Wojewodzic (2011) showed that the number of cattle in the Carpathians decreased significantly in the last decades of the 20th century, mainly due to a lack of profitability, increased sanitary requirements in milk production and an increase in non-agricultural employment. Following the collapse of communism in Poland, most farms in the Polish Carpathians reported a significant decrease in the use of mineral fertilisers and pesticides (Górka et al. 2002). The use of mineral fertilisers in the Carpathians continued to decline after 2000 (Piwowar 2022); this is contrary to development trends observed in other regions of Poland.

Another important factor is the reduction of summer precipitation, which leads to soil desiccation and reduced water retention in the catchment, thus reducing the importance of surface runoff and floods as sources of nutrient inputs (Bochenek 2020). Studies using SWAT modelling in the Bystrzanka catchment showed that if LULC had remained unchanged since 1997, simulated water runoff and nitrogen loads would be 16% and 67% higher than the observed values, respectively (Kijowska-Strugała, Bochenek 2023). Other studies in the Carpathians have also shown that land use has a significant influence on the concentration and load of certain elements, such as phosphorus. Siwek et al. (2013) showed that the PO_4^{3-} content in a forested catchment was several times lower than in an agricultural catchment. These results highlight the significant influence of LULC on nutrient concentrations in streamwater.

This study also revealed higher mean monthly Cl^- values in cold seasons compared with warm seasons. One of the most important non-natural sources of Cl^- is human activity during the winter months due to the use of de-icing salts (Płaczkowska et al. 2024). Data from the District Road Administration in Gorlice (2024) and the Gorlice Municipal Office (2024) showed that the following mixture is used for de-icing roads in the study area: 75% stone aggregate (washed sand of grain size up to 2 mm) or ground slag and 25% salt (e.g., 97% NaCl, 2.5% $CaCl_2$, 0.2% $K_4Fe\ CN_6$). This study also showed that there was a statistically significant negative trend in annual SF and snow cover length during the study period. However, it should be noted that there were years when the snow cover lasted continuously for up to 2 months (e.g., 2010 and 2017). In these years, the average annual Cl^- load was 51.1 kg $\cdot$ ha^{-1} and 31.8 kg $\cdot$ ha^{-1}, which were 80% and 12% higher than the multi-year average, respectively. These de-icing salts were discharged into the stream as a result of surface runoff.

In contrast to lowland areas, mechanical denudation predominates over chemical denudation in mountainous areas (Caine 2004, Beylich, Laute 2021), except for karst regions (Pulina 1974). In the Bystrzanka catchment, the ratio of SSY to DY_R was greater than 1 in 16 years, confirming the dominance of mechanical denudation over chemical denudation. Furthermore, the analysis showed that there was a statistically significant increase in the ratio of SSY to DY_R when the total runoff above the flood threshold increased, once again highlighting the important role of floods in mechanical denudation.

In the Bystrzanka catchment, SSY was 61% higher in the first decade of the multi-year study period than in the last decade. However, Kijowska-Strugała (2019) showed that SSY in 2010–2017 was 7% higher than in 1970–1979, mainly due to the occurrence of an extreme flood in 2010 as well as engineering works in and around the channel. The highest SSY values in mountainous areas are recorded during floods (Lenzi, Marchi 2000, Lana-Renault et al. 2007). Studies have shown that up to 96% of the annual SSL load can be carried away in a single flood (Kijowska-Strugała 2015). High-energy, geomorphologically active floods cause increased erosion and mobilise material for transport. In the Beskids part of the Polish Carpathians, the rate of channel deepening can range between 1 cm · a^{-1} and 8 cm · a^{-1} (Froehlich 1980, Kijowska-Strugała, Bucała-Hrabia 2019), and it is 1.3–3.8 m over the past century (Lach, Wyżga 2002, Krzemień 2003, Wyżga et al. 2016). The mean annual rate of bank erosion ranges between 0.4 m · a^{-1} and 0.6 m · a^{-1} (Tekielak et al. 2007). Many researchers also highlight roads as a source of material transported into the stream channel (Froehlich, Walling 1997, Motha et al. 2004, Cendrero et al. 2022). The sudden increase in catastrophic floods and the acceleration of erosion are directly linked to the rapid runoff of water along low-permeability field roads (Figuła 1966). Experimental studies carried out in the Bystrzanka catchment during heavy rainfall in 2014 showed that the SSC values measured on unpaved roads were up to 96% higher than the SSCs recorded in the stream at the same time (e.g. 16 May 2014; Fig. 12B). Studies in the Carpathian Mountains have shown that about 70–80% of the mean annual suspended sediment removed from the catchment can be attributed to unpaved roads, with an average road dredging rate of 6.6 mm · a^{-1} (Froehlich, Walling 1997). Human activities often interfere with the natural conditions required for the delivery of material to the river channel. Studies by Gil (1976) and Welc (1985) show that, during runoff, about 20% of the material transported into the watercourse comes from the slopes, assuming that two-thirds of the catchment area comprises agricultural land.

Conclusion

This study, conducted in the small Carpathian catchment between 1995 and 2023, highlights the key role that climate change and its associated hydrological changes play in the transport and dynamics of dissolved and suspended matter. The recorded increase in air temperature (0.7°C per decade), which is particularly pronounced in the winter season, translates into a significant decrease in annual SF totals (29.1 mm per decade) and a severe reduction in snow cover duration (16 days per decade). These factors altered the seasonal distribution of runoff and the intensity of denudation processes. The simultaneous prolonged periods of hydrological drought suggest variability in the hydrological regime.

Research on chemical and mechanical denudation in the Bystrzanka catchment has revealed significant changes in the transport of dissolved and suspended sediment; this is due to both natural and anthropogenic factors, including a decrease in cultivated land and an increase in meadow and forested areas. This changed the water balance of the catchment and surface runoff, reducing soil erosion and, consequently, the amount of sediment transported by the river. A decrease in dissolved ion concentrations in the stream indicates a reduction in the intensity of chemical denudation processes in the catchment. Changes in ion concentrations in precipitation were more varied.

Reduced anthropogenic pollution pressures, including reduced fertiliser use and improved wastewater management, have played a significant role in reducing nutrient loads in the stream (38% less in the last decade than in 1995–2004). The 23% decrease in chemical denudation balance over the last year was primarily due to changes in ion concentrations in streamwater. In addition, over the last decade, there has been a 50% decrease in SSL compared with 1995–2004; this has been influenced by several factors, including a reduction in slope erosion as a result of land use changes. Mechanical denudation remains strongly linked to extreme hydrometeorological events, with the highest sediment transport recorded during intense floods. Sediment during flood events is mainly derived from channel erosion and roads.

It should be emphasised that in mountainous areas, chemical and mechanical denudation processes remain critical issues, particularly in light of ongoing climate change and evolving anthropogenic pressures. This study shows that continuous monitoring allows for a detailed analysis of the intensity and direction of long-term changes in mountainous landscapes.

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Author's contribution

Conceptualization and methodology : MKS, WB, SW; validation: MKS, WB; formal analysis: MKS, WB, SW; investigation: MKS, WB, SW; resources: MKS, WB, SW; data curation: MKS, WB, SW; writing – original draft preparation: MKS, WB, SW; writing – review and editing: MKS, WB, SW; visualisation: MKS, WB; supervision: MKS.

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