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Trends in Atmospheric Nitrogen Deposition in Macedonia Studied by Using the Moss Biomonitoring Technique

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Abstract: This study examined the nitrogen content in moss samples collected across Macedonia over a 15-year period (2005, 2010, 2015 and 2020) from 72 consistent sampling locations. The nitrogen content was determined using the Kjeldahl method, providing insight into the trends of atmospheric nitrogen deposition across different regions. Descriptive statistical analyses, including spatial distribution maps, were used to compare the temporal variations and regional nitrogen levels. In addition, box-plots (P_{25}–P_{75}) and whiskers (P_5–P_{95}) were constructed to provide a comprehensive view of the variability across different tectonic units and zones, allowing for an in-depth understanding of the spatial distribution of nitrogen across the country. The study revealed that the median nitrogen content in moss samples decreased from 1.21% in 2005 to 1.04% in 2015, followed by a slight increase to 1.07% in 2020. The highest nitrogen concentrations were consistently found in areas with heavy agricultural activities and high traffic volumes, indicating the direct impact of these anthropogenic factors. The comparisons across regions and geological zones also highlighted the substantial variation in nitrogen levels, reflecting the diverse environmental pressures in different parts of Macedonia. This long-term analysis not only offers valuable insights into the trends in nitrogen pollution but also underlines the necessity for targeted policy interventions, particularly in the regions where nitrogen levels remain persistently high.

Keywords: nitrogen; air pollution; moss biomonitoring; Kjeldahl method; Macedonia

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1. Introduction

Nitrogen pollution is a growing global problem. Excessive nitrogen from agricultural runoff, industrial activities and the burning of fossil fuels has led to significant environmental problems worldwide. In the Western Balkans, industrial activities, energy production and transport are major contributors to nitrogen pollution. Stricter regulations and the increased investment in cleaner technologies are needed to tackle this pollution [1].

Nitrogen is essential for plant growth and is a major component of agricultural fertilisers. However, when nitrogen compounds enter water bodies, they can cause eutrophication, lower oxygen levels and harm aquatic life. In the atmosphere, nitrogen oxides contribute to air pollution by forming smog and acid rain, which can damage ecosystems, buildings and human health [2–5].

Efforts to mitigate nitrogen pollution include optimising the use of fertilisers in agriculture, improving wastewater treatment and reducing the emissions from vehicles and industrial processes. Tackling nitrogen pollution is crucial for protecting biodiversity, ensuring clean water and maintaining healthy ecosystems [1].

Nitrogen pollution has increased significantly over the last century due to human activities. In Europe, the reactive nitrogen emissions are regulated by several European Union directives, such as the National Emission Ceilings Directive and the Nitrates Directive and by the protocols of the Convention on Long-Range Transboundary Air Pollution (LRTAP), including the Gothenburg Protocol. The European Monitoring and Evaluation Programme (EMEP) collects emission data to model the atmospheric transport and deposition of air pollutants using the EMEP Unified Eulerian chemical transport model with a grid size of 50 km $\times$ 50 km [6,7].

The most important anthropogenic sources of oxidised nitrogen include transport, industry and energy production, which account for up to 70% of emissions [8]. The emissions from the soil are also significant, especially under the conditions of high nitrogen input. Lower nitrogen emissions are mainly caused by agricultural activities such as animal farming and fertilisation. Forest fires, which are exacerbated by climate change and human activities, are another important source of nitrogen emissions [9].

The nitrogen cycle, which is significantly disrupted by human activities, has more influence on global ecosystems than any other element cycle. It influences the global climate, ocean dead zones, ocean acidification, stratospheric ozone depletion, biodiversity loss, water eutrophication, groundwater pollution and ground-level ozone pollution. Rockström has identified the nitrogen cycle as one of the nine critical 'planetary boundaries' that need to be managed to avoid the destabilisation of the living conditions [10]. They argue that the nitrogen cycle is already disturbed beyond safe limits.

Biomonitors such as lichens, mosses and certain plant species allow an integrative assessment of ecosystem responses to nitrogen pollution. These organisms can accumulate nitrogen compounds and thus reflect the physiological, ecological and atmospheric conditions of their environment. Monitoring these bioindicators allows researchers to track pollution levels and their impact on ecosystems, which contributes to the development of effective pollution control strategies [11–15].

Passive biomonitoring with mosses could improve the spatial resolution of the nitrogen deposition measurements [15]. The European moss survey, which is carried out every five years, has shown a strong correlation between the nitrogen concentrations in mosses and atmospheric nitrogen deposition rates. The 2005/2006 survey aimed to determine whether mosses can monitor the atmospheric nitrogen deposition across Europe, identify polluted areas and produce European maps [15,16]. The lowest nitrogen concentrations in mosses were found in northern Finland and parts of the United Kingdom, while the highest concentrations were found in western, central and eastern Europe.

Macedonia, a participant in the European Moss Study, has analysed atmospheric nitrogen deposition using moss biomonitoring techniques. The results were compared with those from other European studies. The main aim of the study from 2005 to 2020 was to obtain spatial information on the nitrogen concentrations in the mosses across the country and to identify the polluted areas. Building on previous studies from 2005 to 2010, which marked the country's first steps in monitoring the atmospheric nitrogen levels, the study running until 2020 aimed to provide a more comprehensive spatial analysis of the nitrogen concentrations in mosses [17]. The aim was not only to map the nitrogen deposition across the country, but also to identify and assess the regions with particularly high nitrogen pollution. This extended timeframe provides a clearer picture of the pollution trends and highlights the areas that require further action.

2. Materials and Methods

2.1. Study Area

Macedonia is a landlocked country in the central part of the Balkan Peninsula, located between latitudes 40°50′ and 42°20′ N and longitudes 20°27′ and 23°05′ E. The country covers an area of 25,713 km^2 and has a resident population of 1,836,713 people (according to the 2021 census). Geographically, the country is characterised by a central valley of the river Vardar, which is bordered by mountain ranges. The landscape is predominantly rugged and lies between the Šar and Osogovo mountains, which frame the valley of the Vardar River. In the country, there are three large lakes—Lake Ohrid, Lake Prespa and Lake Dojran—located along its southern borders, which it shares with Albania and Greece. The country is rich in minerals and has significant deposits of chromium, copper, lead, zinc, manganese and nickel [18,19].

Due to its unique natural and geographical features, Macedonia has two different climatic zones: a modified Mediterranean climate and a temperate continental climate. This results in two main seasons: cold, wet winters and dry, hot summers, with transitional periods in the spring and autumn. In the mountainous regions, the climate is characterised by short, cool summers and cold, moderately humid winters, with precipitation falling mainly in the form of snow [20]. The temperate continental climate, an Eastern European subtype, is characterised by cold winters with extended periods of ice, numerous frost days, average monthly temperatures below zero in the coldest months, low average minimum temperatures, low absolute lows and less precipitation. Compared to the Mediterranean climate, the growing season is shorter, humidity is lower, and there are fewer sunny, hot days.

2.2. Sampling

In August 2005, samples of the terrestrial mosses *Homolothecium lutescens* and *Hypnum cupressiforme* were collected from 72 sites evenly distributed across the country, following the sampling strategy of the European Moss Survey Programme (Figure 1). The 2010, 2015 and 2020 moss surveys were conducted in August and September, again collecting 72 samples of *Homolothecium lutescens* and *Hypnum cupressiforme* across the country. Using the same sampling grid as in previous surveys, approximately five to ten subsamples were collected within a 50 m × 50 m area and pooled into a single sample to ensure that the moss samples were representative of each site.

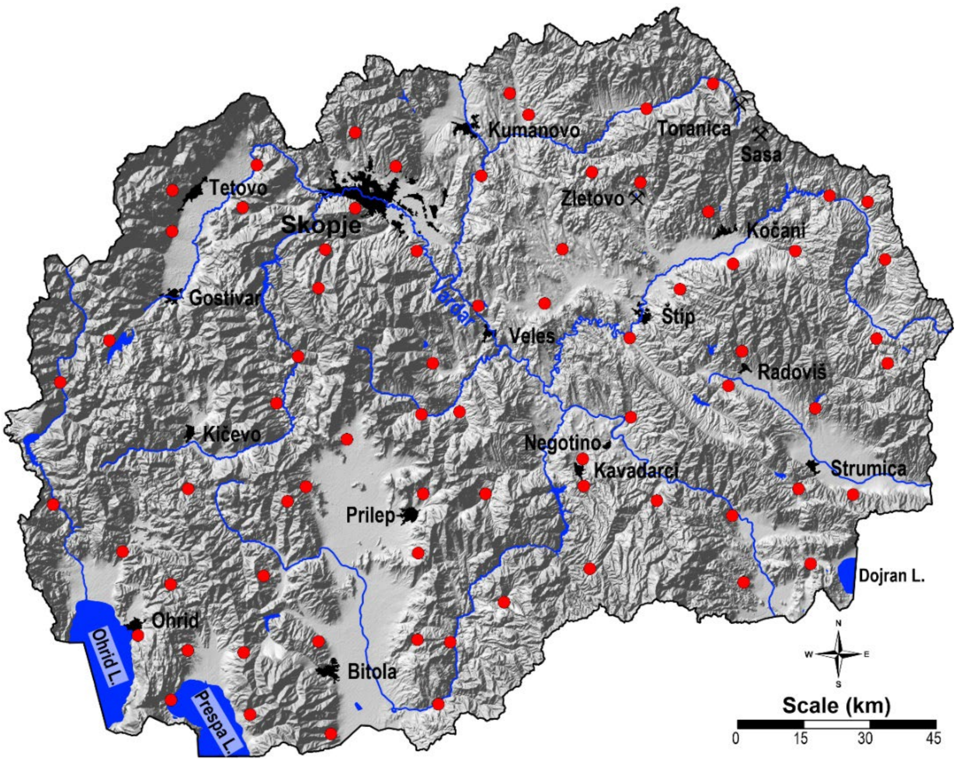

Figure 1. Shaded relief map of Macedonia with sampling locations (red dots).

In 2005, 2010, 2015 and 2020, 72 moss samples were collected, although some samples were missing during the nitrogen analysis phase. Therefore, only 57 samples were analysed for nitrogen analysis in 2005, 68 samples in 2010 and 72 samples in 2015 and 2020.

2.3. Sample Preparation and Analysis

The nitrogen content in the moss samples was determined by using the CN 802 analyser (Velp) and the Dumas method (combustion technique). A sample of 150 mg of homogenised moss material was placed in tin foil for analysis. The instrument conditions were set to an oxygen flow rate of 400 mL/O_2/min. Argon was used as the carrier gas, while oxygen (O_2) served as the combustion gas, both with very high purity (99.99%). Quality control was ensured by the use of duplicate samples and certified reference material (CRM), in particular ethylenediaminetetraacetic acid (EDTA) (VELP). The detection limit for total nitrogen (TN) was 0.1 mg N, and the recovery rate from the reference material was over 95%.

2.4. Mapping

The interpretation and visualisation (mapping), of the spatial distribution of the nitrogen content in the mosses in Macedonia was carried out using the universal kriging method with linear interpolation (ARC MAP 9.x). The base size of the grid cells for the interpolation was set to 0.5 km × 0.5 km.

3. Results and Discussion

The descriptive statistics for the nitrogen content in the moss samples from 2005, 2010, 2015 and 2020 are presented in Table 1.

Table 1. Descriptive statistical analysis of the nitrogen results in moss samples collected in 2005/2020.

Year	N	X	Md	X_{BC}	Min	Max	P_{10}	P_{90}	P_{25}	P_{75}	S	CV	MAD	QCD	A	E	A_{BC}	E_{BC}
2005	57	1.21	1.21	1.23	0.70	1.54	0.97	1.43	1.13	1.37	0.18	15	0.14	10	−0.57	0.18	−0.08	−0.54
2010	68	1.08	1.06	1.05	0.68	1.75	0.86	1.29	0.95	1.18	0.20	18	0.14	11	1.07	2.34	−0.02	0.72
2015	72	1.06	1.04	1.04	0.61	1.96	0.84	1.35	0.92	1.15	0.21	20	0.15	11	1.18	3.73	−0.02	1.20
2020	72	1.07	1.07	1.06	0.70	1.75	0.78	1.29	0.96	1.18	0.19	18	0.14	10	0.37	1.39	0.02	0.75

A comparison of the median nitrogen values from 2005, 2010 and 2015 shown in Table 1 reveals a slight downward trend in the nitrogen content of the moss samples taken every five years. However, the samples taken in 2020 show a slight increase in nitrogen content. In 2005, the nitrogen levels ranged from 0.70% to 1.54%, while in 2010 they were between 0.68% and 1.75%. In 2015, the nitrogen content varied between 0.61% and 1.96% and in 2020 between 0.70% and 1.75% in the moss samples collected throughout the territory of Macedonia. Looking more closely at the median values, the nitrogen content decreased from 1.27% in 2005 to 1.07% in 2020 (Figure 2).

Although there were minor differences in the sample numbers (57, 68, 72 and 72) and sampling periods (August in 2005 and August–September in the subsequent years), these variations were unlikely to significantly impact the results. Moss integrates atmospheric nitrogen over extended periods, so slight shifts in the sampling window should not introduce meaningful bias. Additionally, no reports have shown that the moss nitrogen content is consistently higher in August compared to September or other seasons. Overall, the robust sampling methodology and long-term monitoring ensure the reliability of the study's findings.

This is further illustrated in Figure 3, which shows the spatial distribution of nitrogen. In 2005, most of Macedonia's territory is coloured orange-red, indicating a high nitrogen content. However, in 2010, 2015 and 2020, this trend decreases, with the increased nitrogen content mainly concentrated in the northwestern region of the country.

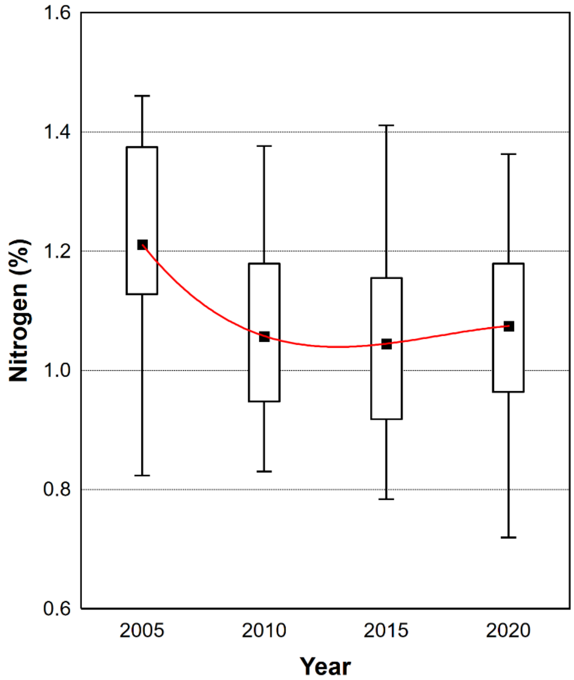

Figure 2. Boxplot analysis of the median nitrogen values from 2005 to 2020, box (P_{25}–P_{75}) and whisker (P_5–P_{95}).

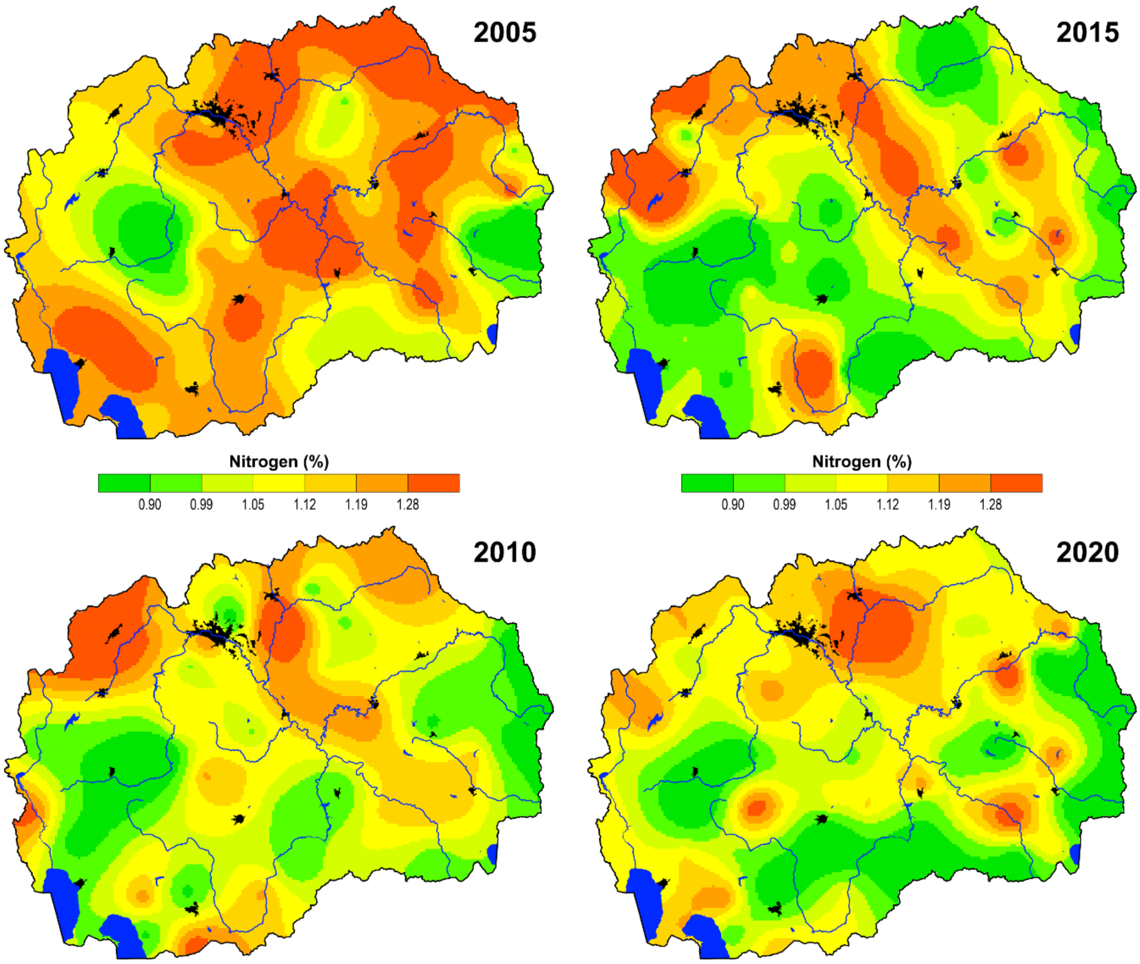

Figure 3. Spatial distribution of nitrogen in moss samples collected in 2005, 2010, 2015 and 2020.

The highest nitrogen content found in the moss samples from 2005 came from a sample taken near the village of Pepelište (central part of the country), where the nitrogen content reached 1.54%. The elevated nitrogen levels in this area can be attributed to the nearby agricultural activities, which include vineyards and vegetable gardens. Agriculture releases reactive nitrogen through the volatilisation and leaching of nitrogen fertilisers [21]. Livestock farming also contributes to the reactive nitrogen emissions through the volatilisation of ammonia gas [22,23]. Industrial activities and motor vehicles also release reactive nitrogen into the atmosphere through the combustion of fossil fuels and other processes that consume large amounts of energy, breaking the triple bond of N_2 and forming nitrogen oxides (NOx, i.e., NO and NO_2) [24]. These nitrogen oxides and ammonia emitted into the atmosphere undergo various chemical reactions that lead to the formation of water-soluble compounds and gases (NO^{3-}, NH^{4+}, HNO_3) as well as aerosols such as ammonium sulphate [$(NH_4)_2SO_4$] and ammonium nitrate (NH_4NO_3) [25]. These compounds are subsequently deposited on the Earth's surface, either as dry deposition—in which atmospheric gases or aerosols settle due to gravity—or as wet deposition, in which nitrogen ions are deposited via fog, snow or precipitation [26,27].

In all the areas of Macedonia, a general trend of decreasing nitrogen levels can be observed from 2005 to 2015, followed by a slight increase or stabilisation in 2020 (Table 2). The initial decrease can be attributed to the improvements in agricultural practices, emission controls and the implementation of environmental measures. However, the slight increase in 2020 indicates that continuous monitoring and the development of new strategies are needed to further reduce the nitrogen deposition and mitigate its impact on the environment.

Table 2. Comparison of nitrogen deposition over a 18-year period, divided by zones and regions, on the territory of Macedonia.

Zone	2005	2010	2015	2020
C_MKD	1.25	1.06	1.10	1.07
E_MKD	1.23	1.05	1.04	1.06
W_MKD	1.20	1.06	1.00	1.05
Ng-Pg	1.22	1.05	1.09	1.08
PM	1.22	1.06	1.00	1.02
VZ	1.23	1.09	1.08	1.09
WMZ	1.22	1.08	1.01	1.09
SMM	1.25	1.00	1.01	1.01

C_MKD—central regions; E_MKD—eastern regions; W_MKD—western regions; Ng-Pg—Paleogene-Neogene basins; WMZ—West-Macedonian zone; PM—Pelagonian Massif; VZ—Vardar zone; SMM—Serbo-Macedonian Massif.

The nitrogen levels in the different regions of Macedonia, measured over a period of 15 years, show subtle fluctuations (Table 2). In 2005, the central region (C_MKD) and the Serbo-Macedonian Massif (SMM) recorded the highest nitrogen levels of 1.25%. The eastern region (E_MKD), the Vardar zone (VZ) and the Pelagonian Massif (PM) followed closely behind, all between 1.22% and 1.23%. Other areas, such as the Ng-Pg Basin (Ng-Pg) and the western region (W_MKD), had slightly lower nitrogen levels, ranging from 1.20% to 1.22%.

By 2010, a gradual decrease in the nitrogen levels was observed in all the zones, with the central region, the western region and the Serbian-Macedonian Massif recording a decrease to 1.06% or less. The Pelagonian Massif recorded a slight increase to 1.09%, the highest value this year. The values continued to fluctuate in 2015, with most regions stabilising at values of between 1.00% and 1.10%. There was a slight increase in the central and eastern regions, while the figure fell to 1.00% in the western region. In the Vardar zone and the Pelagonian Massif, the nitrogen levels fluctuated between 1.08% and 1.09%.

In 2020, the nitrogen levels remained relatively stable with slight changes throughout the region. In the central and eastern region, there was a slight increase to 1.07% and 1.06%, respectively, while the western region remained constant at 1.05%. The Vardar zone and the Pelagonian Massif recorded similar values of 1.09%. These trends suggest that although the nitrogen deposition in Macedonia shows slight fluctuations over time, the overall changes remain within a narrow range, indicating a balanced yet dynamic environmental system across the country.

Statistically, the trends between the different regions and tectonic units did not differ significantly, as shown by the high p-values and non-significant results of the ANOVA tests (Table 3). Significant differences were only observed between different years, which emphasises the importance of the temporal changes over the spatial differences in this context.

Table 3. Comparison of the results of an analysis of variance (ANOVA) from 2005 to 2020 by region and tectonic unit.

Year	Zone	F	p	SIGN
2005–2020	Year	8.11	0.00	*
2005	Region	0.27	0.76	NS
2010	Region	0.05	0.95	NS
2015	Region	1.04	0.36	NS
2020	Region	0.10	0.91	NS
2005	Tectonic unit	0.02	1.00	NS
2010	Tectonic unit	0.57	0.69	NS
2015	Tectonic unit	0.52	0.72	NS
2020	Tectonic unit	0.10	0.91	NS

SIGN: This indicates the significance of the results: * usually denotes statistical significance at a specific level (e.g., $p < 0.05$, $p < 0.01$); NS stands for "Not Significant" meaning the results are not statistically significant at the chosen significance level.

The table contains the results of an analysis of variance (ANOVA), which is a test used to determine the differences between the research results from unrelated samples or groups. The analysis of the nitrogen levels in Macedonia from 2005 to 2020 shows a significant change over time, as shown by the statistical analysis (F = 8.11, p = 0.00). This indicates that the changes in the nitrogen levels over time are significant. In contrast, the spatial variations between the regions in 2005, including the western, central and eastern regions, were not statistically significant (F = 0.27, p = 0.76). This lack of significant regional differences persisted in the subsequent years, including 2010, 2015 and 2020, suggesting that spatial factors do not affect the nitrogen content so significantly.

The correlation matrix of the nitrogen values in Table 4 shows low correlations between the different years, with values of between 0.17 and 0.47. This indicates that the nitrogen values have a certain correlation from year to year, but are not strongly dependent on previous years.

Table 4. The correlation matrix shows the relationships between the nitrogen values over the years.

	2005	2010	2015	2020
2005	1.00			
2010	0.28	1.00		
2015	0.17	0.45	1.00	
2020	0.25	0.47	0.47	1.00

The boxplot analysis of nitrogen values from 2005 to 2020 in Figure 2 illustrates several trends. In 2005, the median nitrogen content was around 1.20%, with an interquartile range (IQR) of 1.10% to 1.30% and a whisker extending from 1.00% to 1.40%. This wider spread and the higher median value indicate the greater variability in the nitrogen load. By 2010, the median had fallen to around 1.05%, with the IQR dropping to 1.00–1.15% and the whisker ranging from 0.80% to 1.30%. This decline indicates a significant decrease in the nitrogen levels and less variability compared to 2005. In 2015, the median nitrogen level stabilised at 1.06%, with the IQR remaining at 1.00–1.15% and whiskers ranging from 0.90% to 1.20%. The data show a narrower range of extreme values, particularly at the lower end. By 2020, the median had risen slightly to 1.07%, with a slightly wider IQR of 1.00–1.20% and whiskers of 0.90% to 1.30%. This indicates a slight increase in the nitrogen levels, although the variability of extreme values has increased compared to 2015.

From 2005 to 2010, the nitrogen levels in Macedonia showed distinct regional patterns, with the western regions initially showing the highest concentrations, around 1.25%. This was likely due to the greater industrial activity or more intensive agricultural practices, as can be seen in Figure 3. In comparison, the central regions followed with values of around 1.23%, while the eastern regions had slightly lower values of around 1.20%. Among the geological zones, the Serbo-Macedonian Massif had the highest nitrogen concentration, with a maximum value of around 1.25%.

By 2010, the nitrogen content in the western regions had fallen significantly to around 1.00%, which represents a dramatic improvement. In contrast, the nitrogen concentrations in the central and eastern regions remained slightly higher, ranging from 1.05% to 1.06%, indicating a gradual decline in pollution. The geological zones such as the Pg-Ng basin and the West-Macedonian zone also recorded decreases, with values dropping to around 1.05% to 1.06%. However, in the Pelagonian Massif and the Vardar zone, there was a slight increase in the nitrogen concentrations to around 1.08% to 1.09%, indicating local factors that may have offset the general downward trend.

The fluctuations in the nitrogen content over the years reveal the notable shifts in the pollution patterns. In 2005, the highest nitrogen concentration was measured in the moss sample from the village of Pepelište (south-central part of the country) with a value of 1.54%, while the samples from the village of Pusta Reka (Skopje region) had the lowest value of only 0.92%. By 2010, the peak nitrogen value had risen significantly. At 1.75%, the moss sample from the village of Belica (central-western part) reached the highest value measured during this period. At the same time, the sample from the village of Zrze (northern part of the Pelagonia region) had the lowest nitrogen concentration of just 0.68%. The fluctuations continued in 2015, when the nitrogen concentration in the sample from the village of Makovo (southern part of the Pelagonia region) reached the highest value so far in Pepelište (1.54%), while the nitrogen content in Belica fell to just 0.61%, which is a stark contrast. In 2020, Zrnovci (central-eastern part of the country) had the highest nitrogen content of 1.52%, while the samples from the two villages of Štavica (near the town of Prilep) and Pekljani (central-eastern part) had the lowest values of 0.70%.

These figures reflect the dynamic nature of the nitrogen pollution in the region and illustrate the influence of the various environmental and anthropogenic factors over time. The trends emphasise the importance of continuous monitoring and adaptive management strategies to mitigate the impact of nitrogen pollution on ecosystems and public health.

When looking at the overall trends from 2005 to 2020, there is a clear decline in the average nitrogen content between 2005 and 2010. This decrease indicates the effectiveness of the initial anti-pollution measures taken during this period. From 2010 onwards, the nitrogen levels stabilised in most regions, although a slight increase was recorded in some areas until 2020. The consistent interquartile range (IQR) observed during these years reflects the stable nitrogen levels, with few extreme pollution events, particularly in 2005 and 2020. This stability suggests that while the pollution control efforts have been largely successful, continued vigilance is needed to maintain these successes and address the new challenges.

The analysis of the nitrogen levels in Macedonia from 2015 to 2020 shows a generally stable trend with some regional variations (Figure 4). In the western regions, the nitrogen content in 2015 was just over 1.05%, the highest among the regions, even though it decreased slightly compared to previous years. By 2020, these values remained constant at around 1.07%, indicating stability in the pollution sources and controls, but also that this region continues to have relatively high nitrogen levels compared to others. In the central regions, the nitrogen levels were around 1.00% in 2015, indicating stability compared to the previous years. This trend continued into 2020, with the levels remaining stable at around 1.05%, indicating that the pollution sources and controls have not changed significantly in this region. In the eastern regions, the nitrogen levels were slightly above 1.00% in 2015, similar to the central regions. In 2020, these values were around 1.05%, reflecting consistency with the central regions and general stability over time.

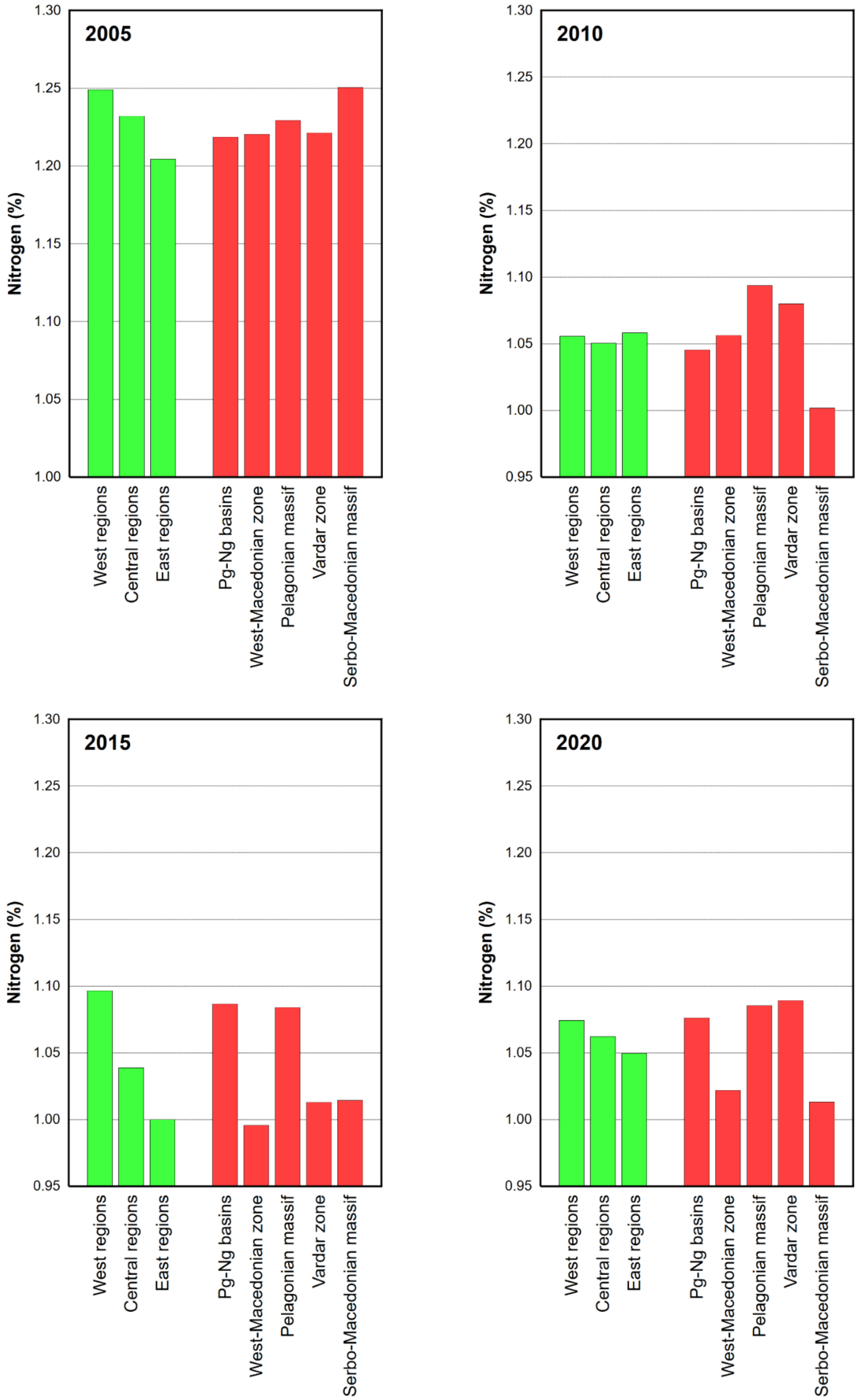

Figure 4. Average nitrogen content in different regions and major geological formations of Macedonia (2005 to 2020).

Looking at the geological zones, the nitrogen levels in the Pg-Ng basin were around 1.08% in 2015, reflecting a decrease from previous years due to the improved pollution control (Figure 4). By 2020, however, the values had risen slightly to just over 1.05%, possibly due to the local changes in the pollution sources.

In the West Macedonian zone, the nitrogen levels were stable at just over 1.00% in 2015 and remained constant at around 1.05% until 2020, indicating effective pollution control measures in this zone. In the Pelagonian Massif, the nitrogen levels were around 1.09% in 2015, slightly lower than in 2010, and had risen to 1.07% by 2020, indicating a possible increase in the pollution sources. In the Vardar zone, the nitrogen levels were 1.05% in 2015 and rose to around 1.08% by 2020, indicating a slight increase in the nitrogen concentrations. In the Serbo-Macedonian Massif, the nitrogen levels were below 1.00% in 2015, which represents a significant decrease compared to the previous years. By 2020, the values remained just below 1.00%, which indicates the success of the measures to combat the pollution in this region.

Overall, the nitrogen levels in Macedonia remained largely stable between 2015 and 2020, indicating effective pollution control. The slight increases observed in the Pelagonian Massif and the Vardar zone indicate the need for continuous monitoring and targeted measures. The persistently higher levels in the western regions emphasise the need for targeted efforts to further reduce the nitrogen pollution in these areas.

Long-Term Distribution of the Nitrogen Levels in Macedonia (2005–2020)

The long-term map of the nitrogen levels in Macedonia (2005–2020) in Figure 5 illustrates the notable variations in the nitrogen concentrations in the country. Through colour coding, where green indicates lower concentrations and orange-red higher values, the map provides valuable insights into the regional and local nitrogen pollution patterns.

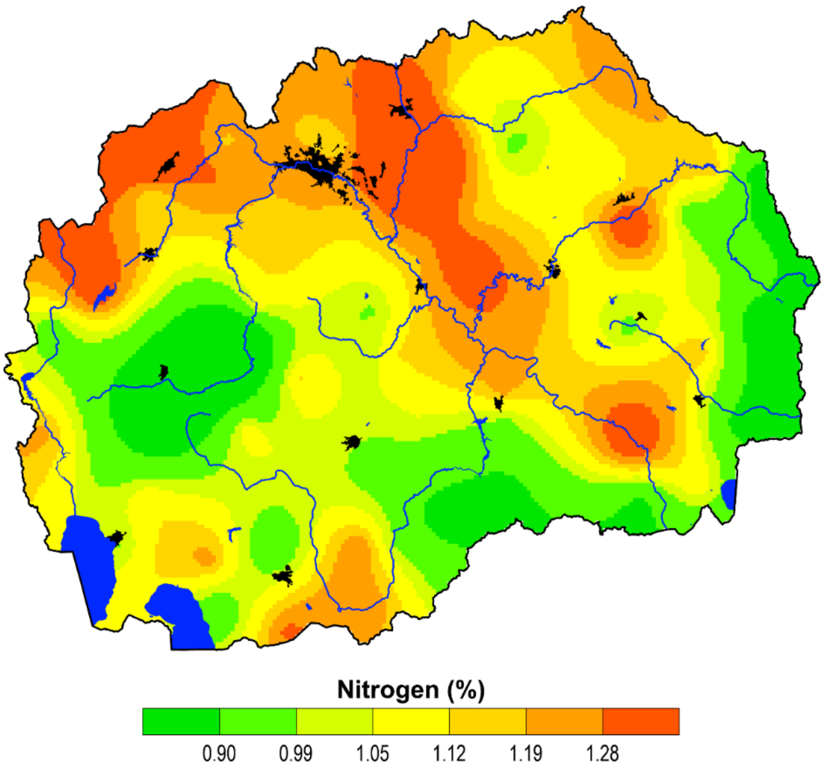

Figure 5. Long-term average nitrogen distribution in Macedonia (2005–2020).

In the northern and northeastern regions, the nitrogen concentrations range between 1.19% and 1.28%, which are consistently the highest values during this period. These increased concentrations are likely to be influenced by the urbanisation leading to an increase in human activities such as transport, construction and energy use, which in turn results in higher nitrogen emissions from fossil fuel combustion. Industrial activities, including the factories and other operations in these regions, also contribute nitrogenous pollutants to the atmosphere and soil. In addition, the heavy use of nitrogen fertilisers in agriculture, especially in densely farmed areas, increases the nitrogen runoff into the environment, exacerbating the pollution. In addition, certain areas in the northwest and Middle East have high nitrogen concentrations, indicating the local pollution hotspots caused by the intensive agriculture and certain industrial activities that contribute to the increased nitrogen levels.

In contrast, the central and southern regions have moderate nitrogen concentrations of between 1.05% and 1.19%. The factors contributing to these values include the agricultural runoff, as nitrogen fertilisers are used less intensively than in the northern regions, but still play a role. The moderate industrial activity also has an impact on these areas, although it is less extensive compared to the northern regions. In addition, natural factors such as precipitation, soil types and vegetation can influence the storage and distribution of the nitrogen in these regions. The eastern regions have similarly moderate nitrogen levels, possibly due to a combination of the environmental factors and land use patterns.

The southwestern and southeastern regions have the lowest nitrogen concentrations, ranging from 0.90% to 0.99%. In these areas, the nitrogen pollution is probably minimised due to the lower population density, which reduces the pollution from transport, industry and human activities. The limited industrial activity in these regions also reduces the nitrogen emissions, while the natural or preserved environments such as the protected areas and forests help to keep the nitrogen levels low. Some central regions also show lower nitrogen concentrations, suggesting the effectiveness of the environmental protection measures or less intensive agricultural activity in these areas.

The spatial distribution of nitrogen content has various effects on the environment. The regions with higher nitrogen concentrations, such as the northern and northeastern areas, may be more susceptible to environmental damage. The potential impacts include soil acidification, which affects the soil health and agricultural productivity, and eutrophication, where increased nitrogen concentrations in water bodies lead to oxygen depletion, damaging aquatic ecosystems. These regions may need targeted policy measures to reduce the nitrogen pollution, including stricter controls on the industrial emissions and improved agricultural practices, such as the promotion of sustainable farming methods to minimise the nitrogen runoff.

On the other hand, the regions with lower nitrogen levels, particularly in the southwest and southeast, should be the focus of conservation efforts to maintain their low pollution status. Conservation measures, such as the promotion of sustainable land use and the expansion of environmental protection, could help to maintain these clean environments.

4. Conclusions

The analysis of the nitrogen content in moss samples collected between 2005 and 2020 throughout the territory of Macedonia shows significant regional differences influenced by industrial activities, agricultural practices and environmental factors. The western regions initially showed the highest nitrogen levels in 2005, probably due to the intensive industrial and agricultural activities. However, by 2010 there was a sharp decline, reflecting the success of the pollution control measures. Improvements were also seen in the central and eastern regions, but these were less dramatic and the nitrogen levels gradually decreased. The geological zones such as the Pg-Ng basin and the West Macedonian zone followed similar trends of decline, although the Pelagonian Massif and the Vardar zone showed a slight increase, highlighting the need for local action.

Individual sites showed significant fluctuations during this period, with sites such as the pastures of Belica, Zrze and Pepelište experiencing notable peaks and troughs. These fluctuations suggest that local factors such as the intensive agriculture or certain industrial activities played a key role in the formation of the pollution hotspots. Despite the general downward trend in the nitrogen levels between 2005 and 2010, there was a stabilisation between 2010 and 2020. While this reflects an improvement, it also emphasises the need for continuous monitoring and adaptive strategies to prevent a possible resurgence of nitrogen pollution.

The results emphasise the importance of taking targeted environmental policy measures, especially in the regions with persistently high nitrogen levels, such as the northern and northeastern regions. At the same time, the regions with lower nitrogen levels, particularly in the southwest and southeast, should continue to be the focus of the environmental protection efforts in order to maintain their lower pollution status. This comprehensive analysis emphasises the need to strike a balance between the economic activity and environmental sustainability and to ensure that the pollution control measures remain effective in both the high- and low-risk regions.

Ultimately, the long-term nitrogen distribution map is an important tool for guiding the future environmental management efforts in Macedonia. It allows policymakers to target the interventions where action is most needed to reduce the nitrogen pollution and protect ecosystems. Through continuous monitoring and adaptive policies, Macedonia can build on the progress of the last decade and protect its environment for future generations.

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