Mineral magnetic properties of sediments in Dragomirna Reservoir, Suceava County, Romania

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ABSTRACT: Sediment cores from Dragomirna Reservoir were extracted in 2012 with two different corers. Downcore susceptibility and remanence of magnetic minerals in the sediment composition were measured to assess their likely sources and pathways of deposition. Variability in the computed magnetic parameters shows the incrementa et decrementa of human footprint on reservoir sediments from 1964 until 2012. This period corresponds to the ascension and decline of industry and agriculture in Suceava metropolitan area. The high sedimentation rate dilutes the magnetic signal in the sediments, but, even under these circumstances, this reservoir proved to be a useful environmental archive.

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KEY WORDS: magnetic minerals, susceptibility, remanence, lake sediments, reservoir.

1. Introduction

Environmental changes affect erosion patterns and sediment transport and deposition and these changes can be recorded by natural archives such as lakes (Dearing et al., 2006). Due to their continuous accumulation over decades to thousands of years (Mîndrescu et al., 2017), lake sediments integrate a wide array of palaeoenvironmental information, and have been extensively used to reconstruct past pollution events (Farmer et al., 1996), changes in land use (Duck and McManus, 1990; Doyen et al., 2016), climate variability (Heiri et al., 2014) and natural disasters, such as floods (Thorndycraft et al., 1998) and volcanic eruptions (McNamara et al., 2018). There are numerous methods used for extracting specific information from sediments, such as the analysis of inorganic geochemical composition (for example by using the X-ray fluorescence (Boyle, 2001)), particle size distribution (Last, 2001), organic and carbonate contents (Heiri et al., 2001), charcoal and fly ash particles (Rose, 2015) or biological remains in sediments such as pollen, plant parts or various fossil crustacean and midges (Smol et al., 2001; 2002). Mineral magnetic properties of sediments were particularly used since 1980s to extract information on sediment sources, type and intensity of catchment erosion, atmospheric pollution or processes within the water body (Sandgren and Snowball, 2002).

Reservoirs are generally much younger water bodies compared to the natural lakes and, therefore, their sedimentary column records only recent Holocene events, i.e., last decades to centuries. This deficiency is compensated by their precisely known age and proximity to areas with dense population and/or intense economic activities, especially urban areas (Shotbolt et al., 2006). This potentially results in a high probability of such archives to record the environmental fingerprint of population and economic development in these areas. Some previous studies (e.g. Butcher et al., 1993) assumed reservoirs are not reliable environmental archives because of their very fluctuant water level; however, more recent studies have demonstrated their usefulness for paleolimnology (Shotbolt et al., 2001). Shotbolt et al. (2005) discussed the main differences in sediment distribution and properties between lakes and reservoirs and highlighted the scientific importance of reservoir sediments when the analysis of reservoir sediments takes into account these differences.

Basic magnetic properties of minerals are key for reconstructing past environmental processes from lacustrine archives, as variations in the type and concentration of magnetic minerals in sediments are closely related to the type and intensity of catchment processes such as soil and bedrock erosion (Thompson and Oldfield, 1986). Minerals that are common in nature, such as quartz, feldspar, calcite, have diamagnetic behavior (very weak negative magnetization when subjected to a magnetic field). They show negative magnetic susceptibility and do not carry remanence. Many natural iron-bearing minerals (such as pyroxene, olivine, garnet, biotite, montmorillonite and carbonates containing Fe and Mn) are paramagnetic at room temperature, i.e., exhibit weak positive magnetization and hence show very low positive magnetic susceptibility and similarly no remanence. On the other hand, Fe, Co, Ni and their alloys show true ferromagnetism (have the highest magnetic susceptibility and remanence). In lake sediments, on catchments with no igneous rocks, these ferromagnetic elements usually have anthropogenic origin. Even if ores of elemental Fe, Co and Ni exist in nature, these are generally oxidized when exposed to the oxygen-rich atmosphere of the Earth and, as a result, produce oxides that could be ferrimagnetic (an example is magnetite). Such ores can be reduced as part of organic matter decomposition (producing sulphides that may be ferrimagnetic, such as greigite) (Sandgren and Snowball, 2002), in sulphur-rich anaerobic environments (such as some lakes). Ferrimagnetic behavior is several orders of magnitude greater than that of paramagnetic and diamagnetic materials. For this reason, ferrimagnetic minerals (such as magnetite) and/or ferromagnetic anthropogenic particles, dominate the magnetic properties of lake-sediments (very high susceptibility and remanence), even if present in low concentrations (Sandgren and Snowball, 2002). Antiferromagnetic minerals have a lower saturation magnetization than ferrimagnetic minerals. Hematite and goethite are the most representative for antiferromagnetic minerals. Hematite is a significant magnetic mineral in oxidized igneous rocks and sediments formed in oxidizing conditions, whereas goethite is a very common mineral, typically formed as a weathering product. The room-temperature saturation magnetization values for all these remanence-carrying minerals (ferro/ferrimagnetic and antiferromagnetic) is known; likewise, the susceptibility of all magnetic minerals, including diamagnetic and paramagnetic minerals, is known. This allows for the mineralogical composition of the sediment, and hence sediment sources and conveying processes to be estimated (Thompson and Oldfield, 1986; Evans and Heller, 2003).

The main scope of this study is to show the temporal variability in mineral magnetic properties of sediments accumulated in the Dragomirna Reservoir (Suceava urban area, north-eastern

Romania), since its formation. Secondarily, we relate the magnetic parameters to sediment geochemistry (from published data – see Briciu (2017)) in order to explain the observed evolutions in the sedimentary column.

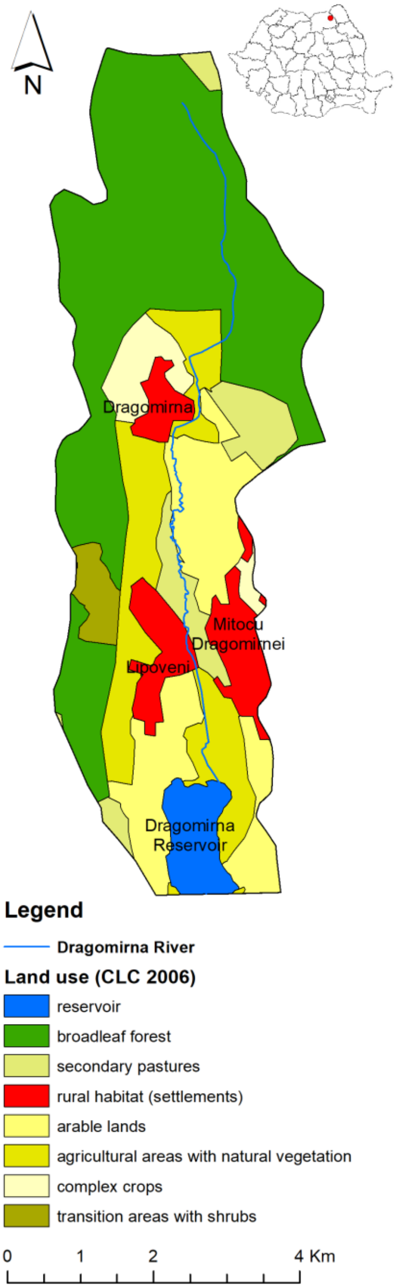

Figure 1 Land use in Dragomirna Reservoir catchment, according to European Environment Agency (2006). This figure is available in colour online at www.georeview.ro.

2. Study area

Ponds and reservoirs are widespread on the territory of Romania, especially in plain and plateau areas (Gâștescu, 1971). First studies concerning these water bodies were focused on bathymetry and water quality, the latter being still of interest today, due to rapid changes in water properties (Romanescu et al., 2014). Although palaeolimological studies on Romanian lakes have expanded in the last decade, no previous attempts have been made to test the use of reservoirs as palaeoenvironmental archives, with the exception, to our knowledge, of Briciu (2017), who described some properties of sediment cores extracted from Dragomirna Reservoir, such as particle size distribution, organic and carbonate content and elemental chemistry.

Dragomirna Reservoir appeared in 1964 behind a dam built on lower Dragomirna River valley (Fig. 1). Dragomirna River is a tributary of Suceava River and has an average flow of approximately 0.08 m^3/s at Lipoveni gauge (Răduianu, 2009), which sometimes exceeded 2 m^3/s at high waters (Briciu, 2017). Dragomirna dam was enlarged and its height raised in 1988. The reservoir belongs to Suceava city administrative area and there is only 1 km distance between the dam and the northwestern limits of the urban built area.

The reservoir usually had bigger areas and volumes during the communist period, when water from Suceava River was pumped into the reservoir for economic purposes (decantation, then removal for consumption – industry, agriculture, population). In the last 10 years, the reservoir usually had areas under 100 hectares and volumes under 10 millions m^3. The sediment origin from the two rivers (Dragomirna and Suceava) is evident in the bimodal distribution of particle sizes in the reservoir sediments (Briciu, 2017).

The water body lies on Volhynian clays and its catchment has a poor diversity of surface rocks. Volhynian rocks are dominant and consist mainly of clays, marls, sands and sandstones (Efros et al., 2004). Pleistocene rocks are found at the southwestern limit of the reservoir catchment and are represented by gravel, sands and loessoid deposits (Martiniuc and Băcăuanu, 1960).

Two distinct socio-economic regimes succeed in Romania between 1964 (when the reservoir was built) and 2012 (when the reservoir sediments were sampled). During the socialist period, Suceava city had important industrial activities, which persisted after the fall of the communism until ca. 1994. Its industries were a heavy source of pollution (AMBRO, IFA, TERMICA). The city underwent strong socio-economic changes during the post-socialist era (Lupchian, 2006), with a sudden shut-down of the heavily polluting industries, but some sources of important air pollution persisted (TERMICA). Remnants of the past air pollution are, without doubt, the high sulfur concentration in the sediments of Dragomirna Reservoir (Briciu, 2017). Most of the catchment is presently used for agricultural purposes (crops and grazing), however such activities were more intensive in the socialist period.

3. Data and methods

Methodology used in this study for sediment magnetic properties analysis is similar to that of Mîndrescu et al. (2013). The reservoir was cored in February 2012, from an ice bridge. An Uwitec gravity corer was used to retrieve the surface sediments and the sediment-water interface, totaling 45 cm of sediment. For the deeper sediments, a modified Russian corer was employed. We obtained a 59 cm sediment core, of which the last 3 cm were bedrock material. The two cores were extracted at ca. 1 m distance, from the central and deeper area of the reservoir.

The cores were sliced at 1 cm intervals, and the samples were weighed, dried at 37°C, ground and homogenized. For determination of magnetic susceptibility, individual samples at every 2 cm along the core were screened with a Bartington Instruments Ltd MS2 meter and MS2B sensor, at both low and high frequencies. Low frequency mass specific (χ) and frequency dependent susceptibility (χFD%) were derived (Dearing, 1999). χ reflects the concentration of strongly magnetic (ferrimagnetic) Fe oxides (magnetite/maghemite), but is also influenced by the presence of paramagnetic and diamagnetic minerals when ferromagnetic minerals are not dominant. χFD indicates the presence of very small, ferromagnetic particles (<0.02μm in diameter) which are superparamagnetic (SPM), produced by pedogenesis and/or burning of soil organic matter. Values of χFD% that are smaller than 2%, mean virtually no SPM grains in the sample (Dearing, 1999).

Properties of magnetic minerals carrying remanence also vary according to their size. Ferrimagnetic and antiferromagnetic minerals with large crystals are structured in sub-regions called domains, and each of these domains can be magnetized in a different direction. These are known as multi-domain magnetic grains (MD) and can lose their acquired magnetization very quickly. Single domain (SD) magnetic grains are small (0.02 - 0.05 μm), more stable and uniformly magnetized. Hence SD grains yield higher magnetic remanence compared to MD grains (Sandgren and Snowball, 2002). Most natural hematite grains are SD, whereas magnetite grains with diameter >1 μm are MD (Thompson and Oldfield, 1986). To test for the magnetic remanence, a Molspin AF Demagnetiser was used for the measurement of Anhysteretic Remanent Magnetisation (ARM), while the Saturated Isothermal Remanent Magnetisation (SIRM) (magnetic field 1.0 T) was transmitted with a Molspin Ltd Pulse Magnetiser. For both parameters, the resulting magnetic remanence was determined with a Minispin Fluxgate Magnetometer (see method in Akinyemi et al., 2013; Hutchinson et al., 2016). Repeated measurements (x3) of each sediment sample were performed, and the resulting values mediated, in order to minimise uncertainties. All obtained values were mass normalised, by dividing the intensity of magnetization to sample dry mass (Walden, 1999). ARM is used to detect SD particles, usually magnetite produced in the pedogenically altered layers, whereas SIRM measures the summed concentration of both SD and MD ferrimagnetic and anti-ferromagnetic minerals. Hence, a ratio between SIRM and ARM indicates magnetic mineral grain size in a sample; samples containing a higher fraction of SD particles will yield lower SIRM/ARM ratios, while samples with increased MD contribution will yield higher ratios. Similarly, a ratio between SIRM and χ would indicate the proportion of paramagnetic minerals (Thompson and Oldfield, 1986; Evans and Heller, 2003).

A principal component analysis (PCA) was employed to explore the relationships between magnetic proxies and elemental composition of sediments. Data for the geochemical elemental composition of Dragomirna Reservoir sediments were taken from Briciu et al. (2017). For PCA, geochemical data was square-root transformed, and the PCA was constructed on the correlation matrix of the data, in PAST (Hammer et al., 2001). For Gravity core, the first principal component (PC1) summed 39.8% of the variance, whereas the second principal component 29.9%. For Russian core, PC1 represented 37% of the variance, and PC2 28.1%.

4. Results and discussion

4.1 Gravity vs. Russian corer

The magnetic and geochemical stratigraphies of the Gravity and Russian sediment cores extracted from Dragomirna Reservoir suggested that the cores overlap almost entirely. That is, the Gravity core comprises 0-45 cm, whereas the Russian core 7-63 cm on the composite depth (Fig. 2; also see Briciu, 2017). Comparison of the two cores also showed a compression / shortening of the sedimentary material in the lower half of the Gravity core (Fig. 2), an issue also highlighted in the literature (e.g., Glew et al., 2002). Given the differences between the two coring devices used (e.g., the Russian corer was originally designed for peat environments), and the reduced thickness of the sediment profile (~60 cm), we chose to present proxy stratigraphies for both cores. These observations are important to take into account in core correlation and construction of the composite sediment profile, especially when different coring equipment is used.

4.2 Downcore variability and interpretation of magnetic parameters

Results show that magnetic parameters vary along the cores, indicating changes in the concentration, mineralogy and grain-size of magnetic minerals. The main magnetic remanence parameters, SIRM and ARM, show pronounced variability in the upper and lower sequences of the sediment profiles, whereas accumulation of magnetic minerals in the middle part of the cores appears relatively stable. Although magnetic susceptibility parameters showed slightly different values between Russian and Gravity cores, the downcore variation of mass-dependent susceptibility (χ) shows in both cases decreases in the concentration of all magnetic minerals in the upper and lower parts of the sediment profile. Frequency dependent susceptibility (χFD%) has in this case very low values, and hence cannot be used as a reliable indicator of topsoil erosion (Dearing, 1999). This suggests that either little topsoil erosion occurred in the lake's catchment, or sedimentation rates were high and diluted the signal, or eroded topsoil magnetic particles did not finally reach the water body.

The top 13 cm of the sediment profile, mostly represented by the Gravity core, show clear decreases and pronounced variations in the magnetic signal. This suggests that some big changes occurred, either natural or anthropogenically induced, on the water body and its catchment. These changes could have been related to lake size variations, and/or changes in sediment sources and erosion intensity. Decreases in SIRM, ARM and χ, together with very high detrital input (Fig. 2), suggest that the magnetic signal was most likely diluted as a result of increased erosion and hence sedimentation rates. An increase in the contribution of MD grains (high SIRM/ARM) and paramagnetic/diamagnetic material (high SIRM/ χ, low SIRM, low positive χ) indicates that the eroded material mostly originated from the bedrock. Clays and clayey marls, which form the lithology of the catchment, generally contain paramagnetic and diamagnetic minerals. Paramagnetic minerals usually have low positive χ values, i.e., lower than 5-10 (10^{-8} m^3 kg^{-1}), whereas diamagnetic minerals show negative χ values, both with no remanence (Dearing, 1999). Lower heavy metal concentration compared to the rest of the sediment profile (Fig. 2; Briciu (2017)) points toward a reduction of atmospheric pollution in this sequence, which also likely contributed to the decrease in the magnetic signal.

Between 13 and 35 cm, there are no major variations in the remanence and susceptibility values. Overall, ARM is slightly higher than the typical values for antiferromagnetic material and low SIRM/ARM indicate significant contributions of SD grains (Thompson and Oldfield, 1986). Thus, a combination of SD hematite and magnetite appears to dominate the magnetic behaviour in this interval. Moderate to high χ, 20-40 (10^{-8} m^3 kg^{-1}), and relatively stable SIRM/χ support this interpretation. Geochemical data show that this interval was characterized by a lower input of detrital material, and high concentration of heavy metals (Zn, Cu) and evaporative Ca (Briciu, 2017). Altogether, this suggests that there were no major interventions on the water body or in the catchment during this time, and that despite relatively high anthropogenic pollution, atmospheric deposition of magnetic particles did not enhance greatly the magnetic signal of the sediments.

The depth interval 35-55 cm is characterized by a maximum SIRM of 200-400 ($10^{-5}Am^2Kg^{-1}$), typical of a combined influence of ferrimagnetic and antiferromagnetic minerals, and decreasing χ with values specific to clay minerals (Thompson and Oldfield, 1986). ARM values are decreasing and slightly higher than those typical of hematite, i.e., ~ 10 ($10^{-5}Am^2Kg^{-1}$). This, coupled with maximum SIRM/ARM, suggests increasing contribution of large MD magnetite grains, which dominate the magnetic signal of the samples. An increasing contribution of detrital input (Fig. 2) in this interval indicates that the hematite was most likely brought by erosion of deeper soil layers, whereas the MD magnetite grains are most likely of anthropogenic origin. Since, as opposed to χ, the SIRM signal is not diluted by the influence of paramagnetic and diamagnetic material, but only by the input of antiferromagnetic minerals, the maximum SIRM in this interval can be interpreted as resulting mostly from an enhanced deposition of anthropogenic magnetic particles.

In the depth interval 55-63 cm, minimum susceptibility and remanence (SIRM is below 200 x $10^{-5}Am^2Kg^{-1}$), coupled with relatively high SIRM/χ and maximum detrital input (Fig. 2) suggest the presence of hematite and goethite, as well as of large amounts of paramagnetic and diamagnetic material which caused a possible dilution of the magnetic signal. The magnetic mineralogy differentiation, coupled with geochemical data, suggests that the haematite & goethite entered the lake basin as products of erosion of deep soil layers and the diamagnetic and paramagnetic minerals likely entered the lake basin as products of bedrock erosion.

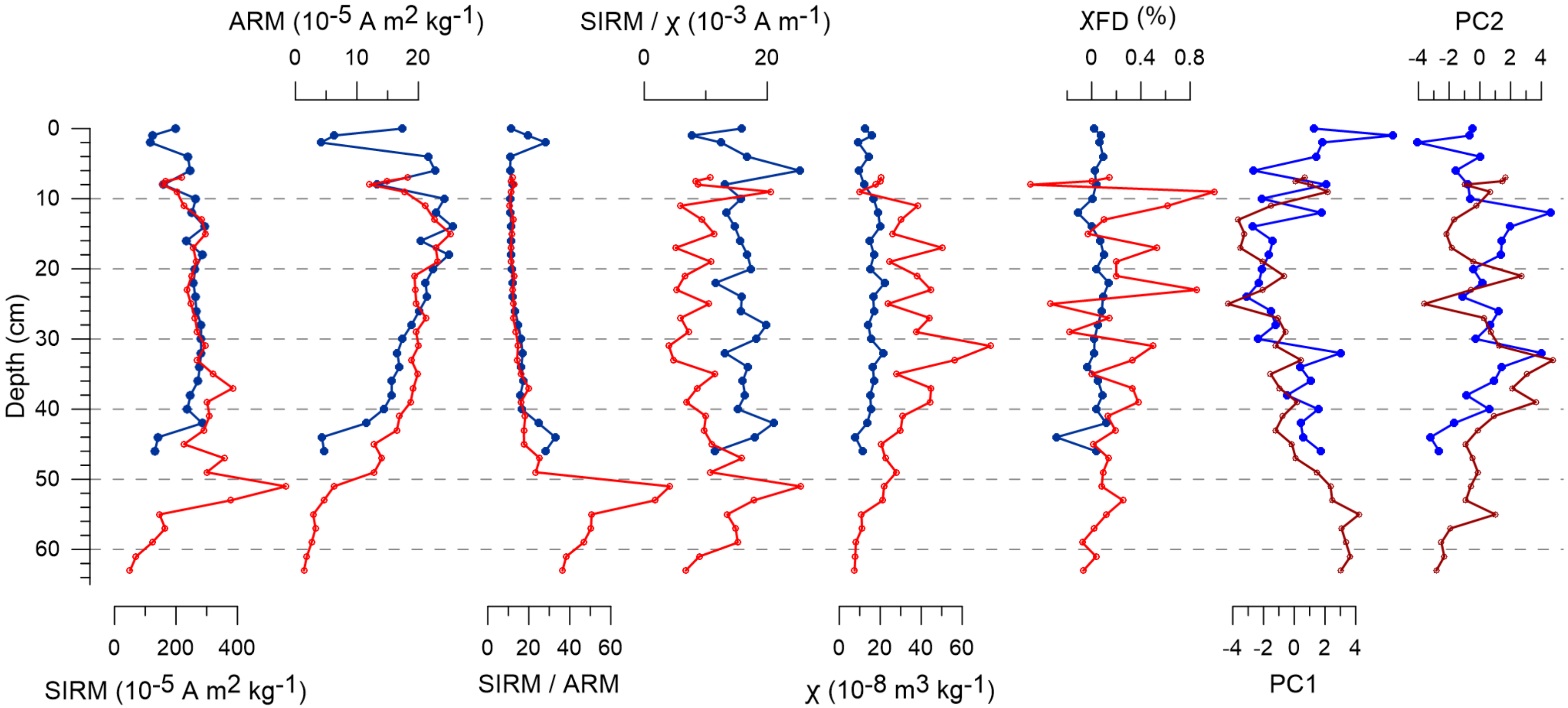

Figure 2 Magnetic remanence and susceptibility parameters and their ratios for Dragomirna Reservoir sediments: saturation isothermal and anhysteretic remanent magnetization (SIRM and ARM), mass-specific and frequency-dependent susceptibility (X and XFD); data for the Gravity core are drawn in blue, whereas for the Russian core in red; PC1 and PC2 are the first two principal components in the principal component analysis (PCA) of the geochemical composition of the Gravity and Russian cores (based on data from Briciu (2017); PC1 sums variance for detrital elements, and PC2 for other geochemical elements, including endogenic elements and heavy metals.

4.3 Spatio-temporal changes in socio-economic activities in the reservoir area

The changes in land use and economic activities over time around the reservoir might be used for explaining the previously discussed downcore variability. There are two socio-economic regimes with distinct characteristics in the lifetime of Dragomirna Reservoir: the communist/socialist regime and the post-communist/capitalist regime. These periods have distinct trends in land use and economy.

Land use around the reservoir during the first period (communist regime, 1964-1989) was characterized by agricultural practices that became more intensive over time (increased use of fertilizers, extended cultivated area), even with irrigated land upstream of Dragomirna Reservoir. That did not lead, most probably, to increased soil erosion, as anti-erosional measures were also implemented. During the second period (1990-2012), such measures were neglected, the agricultural land was divided in more parcels and some parcels abandoned, but the agricultural practices were not intensive. A lower erosion of deeper soil layers and bedrock between 13-55 cm depth, coupled with overall decreased detrital input in this depth interval (Fig. 2) seem to be reflective of these practices. The maximum deposition of bedrock material in the 55-63 cm depth might have originated from the material displaced during the initial construction of the dam in 1964. However, the top 13 cm, characterised by a sharp increase in bedrock erosion, appear to correspond to the beginning of the post-socialist period and might have resulted either from a marked lowering of the water level and/or abandonment of anti-erosional measures and irrigation.

Industry during the first period became more and more pollutant over time, with an apogee in the last decade due to giant factories and plants such as IFA and TERMICA (Briciu, 2017). Only atmospheric pollution, especially atmospheric deposition could have contaminated Dragomirna Reservoir as there were no emissaries from industry into the studied water body. An enhancement with magnetic particles of anthropogenic origin appears evident only in the 13-35 depth interval, as explained largely in the previous section. In the second (post-communist) period, only small factories remained and consistent anti-pollution measures were implemented. This could also explain a decrease in magnetic signal in the top 13 cm of the core (Fig. 2). However, the number of personal cars in Suceava city metropolitan area has followed an impressive upward trend, which has contributed to lower air quality.

Water flow into Dragomirna Reservoir also varied depending on period. There was an increasing trend until 1989 due to increased water volumes pumped from Suceava River into the reservoir. A decreasing trend was recorded from 1990, as fewer water volumes were added from Suceava River and even ceased at the end of the second period due to a severe reduction in industrial activity in the neighbor areas (Briciu, 2017). The progressive increase in SIRM and ARM between 13-35 cm depth is likely reflective of a rather stable water level, whereas the sharp decrease in the top 13 cm might have also been caused by a marked reduction of the lake volume. Water flow from Dragomirna and Suceava rivers (different ratios of these sources over time) might have also affected the variability in sediment mineral magnetic properties.

5. Conclusions

Variability in the concentration and type of magnetic minerals from sediments in Dragomirna Reservoir appear to be a reflection of the past socio-economic developments in Suceava metropolitan area. The most obsolete changes in mineral magnetic properties are shown by SIRM, ARM and their ratio, which have pronounced variability in the upper and lower ends of the sediment profiles. These variations were linked to enhanced input of eroded bedrock material, and to a lesser extent to atmospheric pollution. The measured values of all parameters analyzed in this study might serve as reference for future studies on sediments in Romanian reservoirs.

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