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Influence of hail suppression systems over silver content in the environment in Aragón (Spain). II: Water, sediments and biota

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ABSTRACT

The silver content in soils in the area in the surroundings of the Gallocanta Lake (Aragón, NE Spain) is lower than expected considering the estimated silver emission during the last 50 years by hail suppression systems. To understand the silver accumulation processes, selected water (4 surface, 3 groundwater), biota and sediment (6 surface sediments and 3 cores from Gallocanta and Campillo de Dueñas Lakes and Used Reservoir) samples have been analysed. An essay comparing the growth of wheat in pots with different silver iodide concentrations has been carried out. Finally, silver content in 7 tissues from two sheep that graze in areas both with and without hail suppression systems during 6–8 years were analysed.Our results show that after 50 years of silver iodide emissions to the atmosphere, silver accumulation in the waters and sediments of the lowlands, including some wetlands of high ecological value, has not been significant. Sediment cores did not show any peaks associated to silver emission.We propose that constant absorption of silver by plants could explain the low concentration in water and sediments. Crops and grass could accumulate the excess silver, as both wheat and sheep are able to absorb significant amounts of silver. These bio - accumulation processes could have helped to avoid a progressive environmental deterioration of the surroundings of the Gallocanta Lake. However, this hypothesis should be corroborated and quantified by further research on the analysis of natural and agricultural areas under the influence of hail suppression systems.

1. Introduction

The hail suppression in Aragón (Spain) was initiated in the study area in 1971. Initially, carbon-fueled generators were used, and, from 1973 onwards, ground-based generators that emitted silver iodide dissolved in acetone (BOA, 2016) were installed. After the noon meteorological forecast, the generators were activated by farmers and owners themselves if hailstorm was expected.

Between the 70's and 2001, the Government of Aragón supported hail suppression programmes, such as cloud seeding by aircrafts in 1984 and 1985 (Official Journal of the Aragón Congress, 1985).

In 2001, the next stage was the creation of the Anti-Hail Consortium of Aragón (Official Journal of the Cortes de Aragón, 2001), which gathered the regions usually affected by hailstorms on a voluntary basis. The Anti-Hail Consortium of Aragón installed an initial network formed by 51 ground-based generators (10 × 10 km: Fig. 1), which were operative between April 15th and September 30th. The activation of the generators is based on the daily storm risk (Dessens et al., 2016; Anti-Hail Consortium of Aragón, personal communication), and its functioning is similar to other networks already tested in other areas in Spain (Sánchez et al., 1999).

In the first part of this research (Causapé et al., 2021), silver concentrations in rainfall (5 gauges, 16 samples per site, from April 2017 to March 2019) and soils (72 samples) distributed across the hail suppression network managed by the Anti-hail Consortium of Aragón, were analysed. The results determined the hail suppression systems used in the study area since the early 70's as the source of the silver found in rainfall and soils. The estimation of the silver iodide (AgI) emitted in the surroundings of the Gallocanta Lake during the last fifty years was higher to the silver concentration observed in soils, thus, it would be necessary to go in depth in the study of the accumulation of silver in water, sediments and biota in some places.

Previous studies in Moldova (Potapov et al., 1996), detected lead in water reservoir associated to hail suppression by using lead iodide. However, silver concentration in waters from silver iodide used by hail suppression systems were not significant. In contrast, other studies car-

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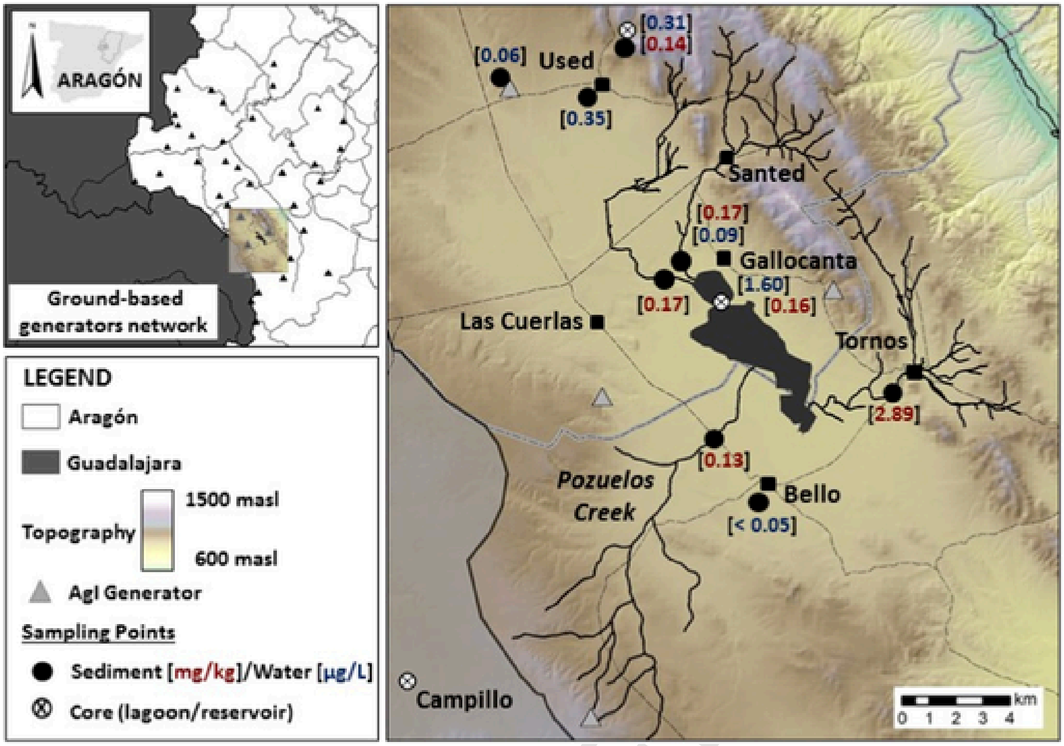

Fig. 1. Location of the study area and silver concentration in water samples, sediment samples and sediment cores.

ried out in the same area, addressed that silver concentration in water reservoirs from protected areas doubled silver concentration in waters from control areas (Cazac et al., 2017).

Silver iodide is insoluble in water, but silver can be dissolved in several ways (WHO, 1996). Silver measured in rivers, lakes and estuaries range between 0.01 μg/L in unpolluted pristine areas, and 0.1 μg/L in urban and industrialised areas (Ratte, 1999).

Depending on the hardness and the salinity of the water, between 30 and 70% of the silver in surface waters are suspended particles (Smith and Carson, 1977). The amount of silver suspended can be high, e.g. silver particles reach 25 mg/kg in the Susquehanna River, which means an estimated flow of 4.5 T of silver to the ocean per year (US EPA, 1980).

Sediments in estuaries where mining waste or sewage are discharged usually have higher silver concentration (>0.1 mg/kg) than sediments unpolluted by anthropogenic activities (Bloom and Crecelius, 1987). Once silver reaches the ocean, it can remain fixed to the sediments up to 100 years (Wingert-Runge and Andren, 1994).

When silver reaches environment, it can also be absorbed by biota. Fajardo et al. (2016) exposed bacteria and Nematoda (edaphic and aquatic biota) to several AgI levels, and they concluded that recurrent cloud seeding over the same area could affect the biota and it is extremely toxic to microflora in aquatic systems (Albright et al., 1972).

Marine algae accumulated silver in water with 2 μg/L of silver, until reaching 58 mg/kg of silver (Sanders et al., 1990). In birds, silver concentration in tissues, especially the liver, were significantly higher in areas polluted by metals in the San Francisco Bay (Szefer et al., 1993).

In spite of the aforementioned bio-accumulation, several studies have stated that silver iodide accumulation is not dangerous for humans (Standler and Vonnegut, 1972: WMA, 2009). However, a well-documented consequence of the excessive silver absorption is algiria, which alters skin and hair colour due to the accumulation of silver in tissues (WHO, 1996).

International directives published by the World Health Organisation (WHO, 2003) or the European Union (EC, 2000; EC, 2006a; EC, 2006b) does not establish thresholds for silver content in water and foodstuff, not even include silver in the list of pollutant to be controlled. At a national level, law about drinking water quality (BOE, 2003) neither mention silver as a pollutant. In fact, salts of silver are used to control bacteria pollution in drinking water, and the threshold for health risk is established in 0.1 mg/L. This threshold is also the secondary standard of silver established by the US Environmental Protection Agency (US EPA, 2015). However, to track mobility of the silver emitted during decades it should be a mandatory environmental goal.

2. Methods

2.1. Water and sediments

Based on the findings of the first part of the research, ten samples in the surroundings of the Gallocanta Lake were collected with the aim of analysing how the silver is transported and accumulated (Fig. 1). The samples were collected in several ecosystems: water from the Gallocanta Lake, creeks that flow to the lake, springs and fountains in some villages.

In seven out of ten sites, water samples were collected, and in six of them, sediments were collected. Additionally, two gravity cores were recovered in the deepest part of the Used Reservoir and the Campillo de Dueñas Lagoon, and one in the middle of the northern part of the Gallocanta Lake. All the cores have the same length (0.3 m), and they were sub-sampled every 0.5 cm the upper 20 cm, and every 1 cm the deeper 10 cm.

Silver content and trace elements in the samples were analysed by Inductively Coupled Plasma-Mass Spectrometry (ICP-MS). Solid samples were previously digested by HF, HNO_3 and $HClO_4$, mixed and concentrated, and the residue was dissolved with HNO_3 6%. The analysis was carried out using the Agilent 7500ce system with ORS (Octopolar Reaction System) technology, which use helium collision mode, and its measurement procedures are based on the U.S. EPA 200.8. (1994)

guidelines for liquids samples and the U.S. EPA 6020B (2014) for solid ones.

The construction of the age-model for Gallocanta Lake was obtained from Burjachs et al. (1996), who determined a sedimentation rate of 0.3 cm/year of the topmost 45 cm by using ^{137}Cs and ^{210}Pb essays. In the Used Reservoir, the bottom of the core reached the most ancient sediments of the reservoir, built in 1983. The sedimentation rate gives a rate of 1.1 cm/year. Finally, the age-model for Campillo de Dueñas Lagoon was obtained by combining ^{14}C, ^{137}Cs y ^{210}Pb essays, and the sedimentation rate provided was similar to the rate in Gallocanta.

2.2. Biota

2.2.1. Wheat

With the aim of analysing the possible silver absorption by the crops in the study area, an essay with five pots planted with wheat was carried out (Table 1). To do so, a range of AgI dissolved in acetone (concentration = 420 g/L) used by the ground-based generators were mixed in 15 kg of soil in each pot.

Given the difficulty of homogeneously mixing AgI in the soil, silver concentrations in the pots were tested in laboratory aiming to check out if the expected range of concentrations had been successfully obtained.

Once wheat grew in the pots, the plants were analysed and compared to samples of wheat and straw collected in the surroundings of the Gallocanta Lake, with the aim of comparing silver concentration and estimating the amount of silver removed by harvests.

Soil samples from the pots were treated by aqua regia and tested by atomic absorption. Wheat and straw samples were analysed and digested with hydrochloric acid and they were also tested by atomic absorption.

2.2.2. Sheep tissues

Based on the idea which stated that silver emitted by the hail suppression systems can be absorbed by herbivorous vertebrates, tissues from several parts of two sheep that had been grazing around 6–8 years both in areas with and without hail suppression systems have been obtained.

The following tissues were sampled: liver, kidney, spleen, fur, hooves, bone, and lean. Silver concentration in the tissues was compared aiming to observe where silver is prone to be accumulated, since this information may be useful in the future for further researches including more vertebrates.

The samples were freeze-dried and calcined up to 500 °C. The ashes were dissolved in hydrochloric acid and the silver content was obtained by atomic absorption.

Table 1
Parameters of the silver concentration essays in pots planted with wheat.

Pot	Soil	Hail suppression product		Soil (theroretical/lab)	
	kg	AgI (mg/L)	ml	Ag (mg/kg)	
1	15	420	0,1	1	4,0
2	15	420	0,5	6	5,5
3	15	420	1,0	13	24,9
4	15	420	3,0	38	71,4
5	15	420	5,0	64	104,8

3. Results

3.1. Water and sediments

Silver content in water samples (Fig. 1; Table 2) are higher than the content naturally found in unpolluted waters (0.01 μg/L; Ratte, 1999), but they are still below the secondary standard established by the US EPA (2015).

Even water from a tank next to a ground-based generator in Used had lower silver concentration (0.06 μg/L), although the tank had been recently refilled when the sample was collected, which could dissolve the concentration.

Silver concentration in the Used Reservoir and in a public fountain in Used, which are respectively feeded by surface and ground waters from Sierra de Santa Cruz, were higher (0.31 and 0.35 μg/L). These concentrations are of the same order of magnitude that drinking water sampled in the village (0.21 μg/L).

However, drinking water in Bello, which already have serious issues related to nitrate pollution from agricultural sources (Orellana-Macías et al., 2020), were below the detection limit (< 0.05 μg/L).

Additionally, freshwater constantly renewed such as the Santed Creek, which comes from the Palaeozoic rocks of the Sierra de Santa Cruz and flow into the Gallocanta Lake, had lower silver concentrations (0.09 μg/L). Silver content in water may be accumulated in the lake, which had the highest concentration of the area (1.6 μg/L). When the samples in the lake were collected, water level was very low, and the extension of the lake was reduced, so evapotranspiration could enhance silver concentration.

Regarding to the analysis of the sediments collected in the creeks of the Gallocanta Basin, the Used Reservoir, and the Gallocanta Lake it, all of them showed higher silver concentrations than soil sampled in the same area (Causapé et al., 2021). Those concentrations were higher than 0.1 mg/kg, the threshold established by Bloom and Crecelius (1987) to classify a soil as polluted by anthropogenic activities. Silver concentration was significantly higher in Tornos Creek (2.89 mg/kg), which may be explained by silver transport through creeks and streams.

In relation to the cores (Fig. 2), silver content in the Gallocanta Lake was below the detection limit (< 0.1 mg/kg) throughout all the sequence, except for the deepest interval (21 and 24 cm depth), where reached 0.14 mg/kg. Silver content in the Campillo de Dueñas core was around the detection limit, but the deepest interval (25 and 27 cm) had higher amounts (0.4 mg/kg and 0.24 mg/kg, respectively). Finally, silver content in the Used Reservoir core was constant, slightly above the detection limit (0.1–0.15 mg/kg).

Table 2
Silver concentration in water and sediments in the surroundings of the Gallocanta Lake.

Sampling point	Water	Sediments
	Ag (μg/L)	Ag (mg/kg)
Tank generator used	0.06	–
Fountain used	0.35	–
Used reservoir	0.31	0.14
Used drinking water	0.21	–
Bello drinking water	< 0.05	–
Santed Creek	0.09	0.17
Used Creek	–	0.17
Tornos Creek	–	2.89
Pozuelos Creek	–	0.13
Gallocanta Lake	1.60	0.16

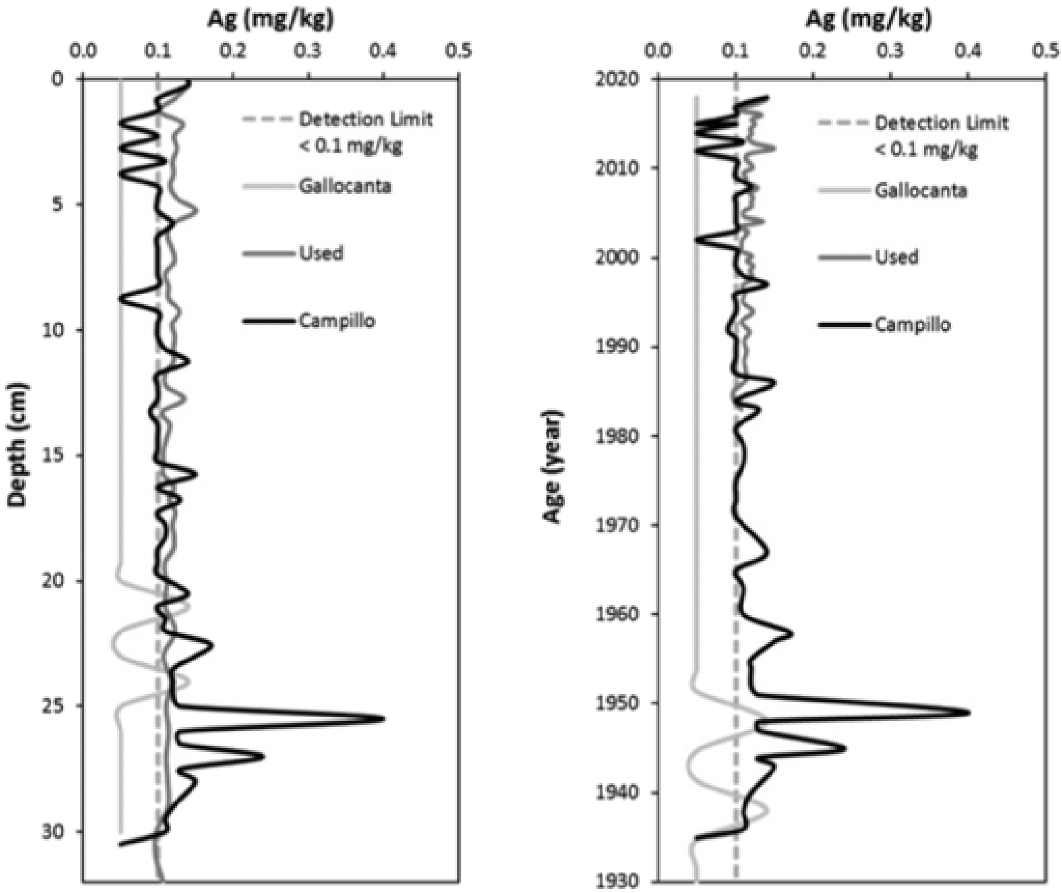

Fig. 2. Silver content in the Gallocanta Lake core, the Used Reservoir core and the Campillo de Dueñas core in the depth sequence (left), and the age-model sequence (right).

The age-model sequence of the cores did not show peaks of silver associated to periods in which hail suppression systems were actively functioning, such as the installation of the firsts ground-based generators (1971), the verified use of aircrafts (1984–1985), or the installation of the Anti-hail Consortium of Aragon network (2001).

Based on the results obtained from the cores, silver content throughout time are in the same order of magnitude that soils in the study area (Causapé et al., 2021), and the spatial variability is low. The Used Reservoir had higher concentration than Campillo de Dueñas, where hail suppression systems are not in use since the late 70's, whereas concentration in Gallocanta Lake were lower. Considering the sedimentation rate of each site (1.1 vs 0.3 cm/year), the amount of silver that reached Used Reservoir was a few time higher than in the Gallocanta Lake and in the Campillo de Dueñas Lagoon.

Two peaks in silver concentration are remarkable in the Campillo de Dueñas Lagoon, in 1945 and 1949, which are correlated with peaks in the Gallocanta Lake (Fig. 2). There is not any sedimentological evidence (i.e. geochemistry, grain size, colour, etc.) which explains a silver movement along the profile. Based on the available information, those peaks cannot be easily explained, since there are no documentary proofs of silver iodide emissions during those years. It would be necessary further research to find correlations between peaks in silver content and other elements.

3.2. Biota

The essay carried out in pots showed that wheat growth was limited when silver concentration in soils increase, to a point where the pots with the highest concentration were not able to grow, as stated by US EPA (1980) about silver effects over germination. However, silver content in soils of the study area was significantly lower, so the estimation of a likely loss of production would need a meticulous agronomic study.

The aforementioned consequences of silver concentration over wheat growth were combined with a higher silver concentration in it straw (Table 3). Silver content in grain could not be analysed since the wheat could not reach that stage. In the case of silver content in wheat sampled in the study area, concentration in straw was 0.19 mg/kg, whereas concentration in grain was 0.15 mg/kg.

Regarding to the sheep tissues, one of them graze in an area under the influence of silver content its whole life. This sheep had higher silver content in all the analysed tissues (Table 4). The highest concentration was found in the liver, which was already reported in other animals (Szefer et al., 1993).

4. Discussion and conclusions

Silver concentrations in waters were above 0.01 μg/L, which is the threshold proposed by Ratte (1999) for pristine areas. However, concentrations are significantly below the secondary standard threshold established by US EPA (2015) for silver (100 μg/L), as the ones obtained in Moldova (Potapov et al., 1996; and Cazac et al., 2017). Nevertheless, Cazac et al. (2017) concluded that silver concentration in water pods in protected areas doubled the concentration of areas without hail suppression systems.

Additionally, silver content in the sediments was slightly higher than those observed by Causapé et al. (2021) in soils of the same area. This suggests that silver is transported and lixiviated by streams, as stated in Smith and Carson (1977).

Table 3
Essays of silver content in soils and straw in pots planted with wheat.

1 mg/kg soil	6 mg/kg soil	13 mg/kg soil	38 mg/kg soil	64 mg/kg soil
				
1,6 mg/kg straw	2,5 mg/kg straw	7,4 mg/kg straw	–	–

Table 4
Silver content in seven tissues of two sheep that graze during 6–8 years both in areas with and without ground-based generators.

	Liver	Kidney	Spleen	Fur	Hoof	Bone	Lean
	Ag (mg/kg dried)						
Hail suppression	10.2	2.7	0.3	1.7	1.0	1.7	1.9
No hail suppression	0.5	0.3	<0.1	<0.1	0.8	1.6	1.9

However, the amount of silver reaching the lowlands of the study area is generally small. Overall, silver concentrations in sediments from the Gallocanta and the Campillo de Dueñas Lakes and the Used Reservoir are around 0.1 mg/kg, which is the threshold proposed by Bloom and Crecelius (1987) for considering an ecosystem as affected by anthropogenic activities.

In addition, silver content in the sediment cores is relatively constant, and according to the age models, peaks of silver do not correlated with more intense periods of hail suppression activities, such as the installation of the first ground-based generators (1971), the use of aircrafts (1984–1985) or the installation of the Anti-Hail Consortium of Aragon network (2001–2017).

The low silver concentrations in nearby soil, sediments and waters could be a reflection of the dispersion of silver particles once they reach the atmosphere. As stated by Freeman (1979) and Jing et al. (2016), particles can be transported hundreds of kilometres away from the sources points, thus, most of the silver can be deposited in areas beyond the target zones. This hypothesis is supported by the higher silver content detected in the Campillo de Dueñas core, located in an upwind area in Guadalajara, where hail suppression stopped in the late 70's.

Regarding to the assessment of the silver bio-accumulation in vertebrates, the analysis of tissues from sheep concluded that silver from hail suppression systems can be bio-accumulated in tissues of herbivores grazing in the area, and it is mainly accumulated in the liver. Our findings are similar to Younger and Crookshank (1977) studies. They expose young sheep to daily silver iodide doses, reached a similar conclusion. Sheep exposed to higher silver iodide doses (168 mg/day during 86 days) accumulated higher silver concentration in tissues, and the liver was the tissue with the highest concentration (17.4 mg/kg).

It is noteworthy to mention that Younger and Crookshank (1977) did not found consequences related to toxicity in their essays, which supports the conclusion reached by Cooper and Jolly (1970), who stated that fishes, microorganisms and invertebrates are more fragile to silver toxicity than humans and vertebrates.

In relation to the essays with pots planted with wheat, they showed that when silver concentration is high enough, it may be toxic for plants, and its growth and productivity can be seriously affected. However, silver concentration observed in the surroundings of the Gallocanta Lake were significantly lower than those required for toxicity impacts.

Silver concentration observed in soils and sediments is significantly lower than expected after 50 years of emission (Causapé et al., 2021). We propose bio-accumulation processes as absorption and subsequent extraction by plants and crops as the main responsible for these lower concentrations.

The experimental plots have demonstrated that wheat is able to absorb silver in relatively high quantities. Given the silver concentration observed in wheat grain (0.15 mg/kg) and in wheat straw (0.19 mg/kg) from the study area, and based on the average harvest in the area (4000 kg/ha and 2000 kg/ha, respectively), the theoretical output would be 0.10 $mg/m^2 \cdot year$. This output is similar to the estimated atmospheric deposition in the study area quantified by Causapé et al. (2021) for the period 2017–2018.

The higher wheat crops accumulation may explain the higher silver content in sediments of the Used Reservoir compared to the Gallocanta Lake. Whereas Used Reservoir is fed by streams from the mountains that flow across natural forested areas, the recharge area of the Gallocanta Lake is mostly extensive cereal croplands. Scrubland and forests are the main land cover in the mountains, which are also able to absorb silver, specially dense and old trees. However, agricultural lands may not only absorb silver, but also remove it from the ecosystem. This also would explain the lower silver concentration in drinking water of Bello, which is already polluted by nitrates lixiviated from agricultural lands, compared to wells fed by waters from the mountains.

In conclusion, low silver concentrations in soils and sediments in the Gallocanta area after several decades of high AgI emissions by hail suppression systems could be explained by preferential Ag absorption by biota, especially cereal crops, and elimination from the ecosystem by harvest.

This hypothesis should be corroborated by further analysis of agricultural and natural lands with the aim of going in depth in the amount of silver that can be absorbed and removed in areas differently affected by hail suppression.

CRediT authorship contribution statement

Jesús Causapé: Conceptualization, Methodology, Formal analysis, Investigation, Writing – original draft, Visualization, Supervision, Project administration. **José María Orellana-Macías:** Formal analysis, Writing – original draft, Writing – review & editing, Visualization. **Blas Valero-Garcés:** Conceptualization, Methodology, Resources, Data curation, Writing – review & editing. **Iciar Vázquez:** Data curation, Writing – review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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