

## Supplementary S2. Lake Oltina chronology

A chronology for Lake Oltina was established on the basis of 17 AMS  $^{14}\text{C}$  measurements (Table 1a, b). Attempts to constrain the chronology of the top core via  $^{210}\text{Pb}$  and  $^{137}\text{Cs}$  gamma assay measurements failed to produce any meaningful results probably due to sediment mixing. An attempt via the geochemical identification of tephra also failed due to a poor match to any known volcanic eruption.

The radiocarbon age estimates were converted into calendar years BP via BACON software (Blaauw and Christen 2011) using the INTCAL13 data set of Reimer et al. (2013). An age–depth curve was derived based on a smoothing spline model. Calendar age point estimates for depths were based on weighted average age–depth curves and also by taking into account the error range of the calibrated ages. Due to the lack of plant macrofossils, we used mostly shells and bulk material for the radiocarbon measurements. Consequently, age determination was problematic due to the low organic carbon levels and possible hard water effects (Table 1b).

We have attempted to correct for the reservoir effect in the following ways. Firstly, we compared our youngest radiocarbon date on the shell sample from 30 cm in depth (880 uncal. Yr BP) with the potential sediment age of recent samples based on geochemistry, mineral magnetic measurements and specific pollen marker. From 30 cm geochemical elements (particularly Pb) potentially associated with regional industrialisation (post-1850) show concentrations above the background levels that would naturally be found suggesting an additional anthropogenic input (Fig. 1). Further, mineral magnetic properties (X) also show an increase from 30 cm that might reflect an anthropogenic influence on the sediment (Fig. 1). Rose et al. (2009), Akinyemi et al. (2013) and Hutchinson et al. (2016) note in the Romanian Carpathian Mountains atmospherically derived inputs of trace elements and heavy metals and mineral magnetic particles from the start of the 20<sup>th</sup> century with peaks from the 1950s. Similarly, Begy et al. (2012) attribute peaks in heavy metals in a lake in the Danube delta to industrial and traffic pollution from the 1950s. We noted the occurrence of pollen of *Ambrosia*, an invasive species that arrived and spread post 1850, also increased at this depth (Fig. 1). Taken together, results from the geochemistry, mineral magnetic measurements and pollen agree in suggesting that the age of the sediment at 30 cm must be post-1850 AD. Based on the difference of the two age estimates (880 years on the shell) and ~50-100 years (via the geochemistry, mineral magnetic measurement and pollen), the age offset at 30 cm is about 700 years.

Secondly, for older sediments, we compared the radiocarbon date of the terrestrial macrofossil sample at a depth of 548 cm (3459±3) with the shells from the same layer (4457±35). Here the age difference between the two measurements is 1000 years, which is close to the 700 years age offset observed at 30 cm. Consequently, we have estimated the hard water effect at about 800 ± 200 yrs. In the Bacon model an age offset of 1000 years was specified for all radiocarbon dating on shells (Fig. 2). Furthermore, all measurements performed on bulk samples were rejected from the age–depth model. This is because the reservoir effect on bulk sediment is much larger than for shell, with a much larger possible error. In addition, in at least two age measurements on bulk samples (at 150 and 230 cm) the H-fraction was very different from L fraction, which indicates that the bulk organic matter is a composite of very different age organic material. The results of our final model provide an age–depth curve with fewer age depth reversals than seen when including the bulk samples.

Table 1a. AMS  $^{14}\text{C}$  measurements at Lake Oltina.

Lab. no.	Core	Depth (cm)	Material	$^{14}\text{C}$ age (( $\pm 1\sigma$ ))	Obs
DeA-10928	1.1	20 (30)	Shell	880 $\pm$ 19	
DeA-11083	1.2	95 (105)	Bulk	2053 $\pm$ 28outlier	
DeA-11085	1.2	150 (160)	Bulk	1832 $\pm$ 27outlier	
DeA-11087	1.3	230 (240)	Bulk	1928 $\pm$ 37outlier	
RoAMS 131.45	1.3	270 (280)	Shell	3016 $\pm$ 30	
RoAMS 128.45	1.4	364 (374)	Shell	3473 $\pm$ 23	
RoAMS 132.45	1.5	442 (447)	Shell	4042 $\pm$ 24	
RoAMS 366.45	1.5	482 (487)	Shell	4715 $\pm$ 34	
RoAMS 133.45	1.6	548 (548)	Plant	3459 $\pm$ 31	
RoAMS 356.45	1.6	547 (547)	Shell	4457 $\pm$ 35	
RoAMS 134.45	1.7	630 (620)	Shell	4856 $\pm$ 25	
RoAMS 364.45	1.7	693(5) (683)	Shell	5097 $\pm$ 36	
RoAMS 129.45	1.8	769 (752)	Shell	5476 $\pm$ 25	
RoAMS 135.45	1.9	826 (804)	Shell	5648 $\pm$ 27	
RoAMS 130.45	1.10	920 (891)	Shell	5055 $\pm$ 30	
RoAMS 363.45	1.10	955 (926)	Bulk	8886 $\pm$ 41outlier	
RoAMS 361.45	1.10	992 (963)	Shell	6093 $\pm$ 42	

Table 1b. AMS  $^{14}\text{C}$  measurements at Lake Oltina showing different age of the H-and L-fractions of bulk samples for two of the four samples performed using bulk material.

AMS $^{14}\text{C}$ Lab Code	HEKAL Sample Nr.	Sample name (sample material dated)	Conventional $^{14}\text{C}$ age (yr BP) ( $\pm 1\sigma$ )
DeA-10928	I/1451/1	Lake Oltina_20, Romania (fragment of large shell)	880 $\pm$ 19
DeA-11083	I/1451/2L	Lake Oltina_95, Romania (bulk)	2053 $\pm$ 28
DeA-11084	I/1451/2H	Lake Oltina_95, Romania (bulk)	2818 $\pm$ 42
DeA-11085	I/1451/3L	Lake Oltina_150, Romania (bulk)	1832 $\pm$ 27
DeA-11086	I/1451/3H	Lake Oltina_150, Romania (bulk)	2374 $\pm$ 27
DeA-11087	I/1451/4L	Lake Oltina_230, Romania (bulk)	1928 $\pm$ 37
DeA-11088	I/1451/4H	Lake Oltina_230, Romania (bulk)	2013 $\pm$ 24

Figure1. Selected geochemical elements (Pb, Zn), mineral magnetic measurements (magnetic susceptibility) and pollen (*Ambrosia*) displaying their simultaneous increase after 30 cm (vertical dashed line) reflecting the post 1850 AD trend of regional industrialisation and the known spread of an invasive weed.

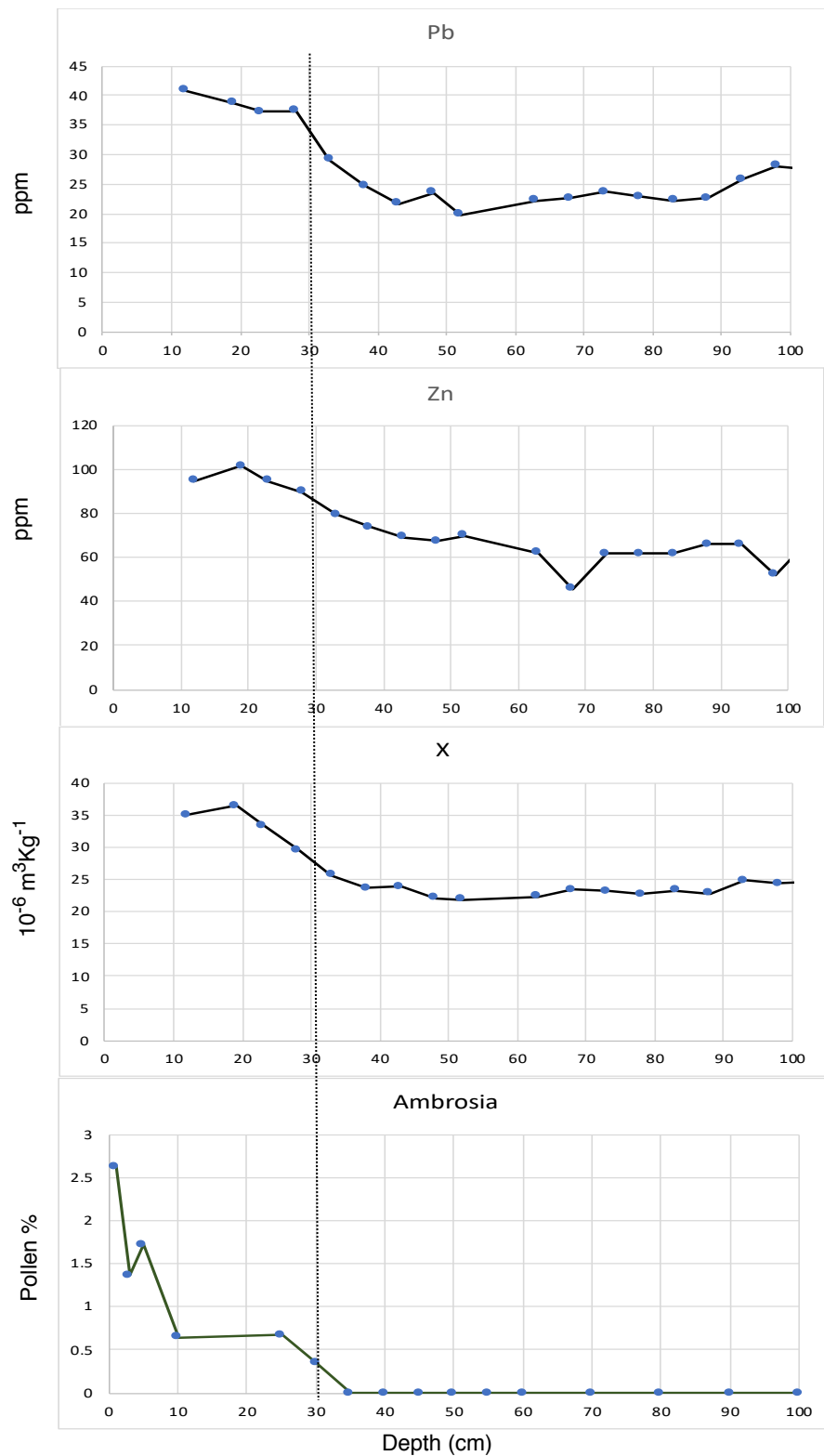
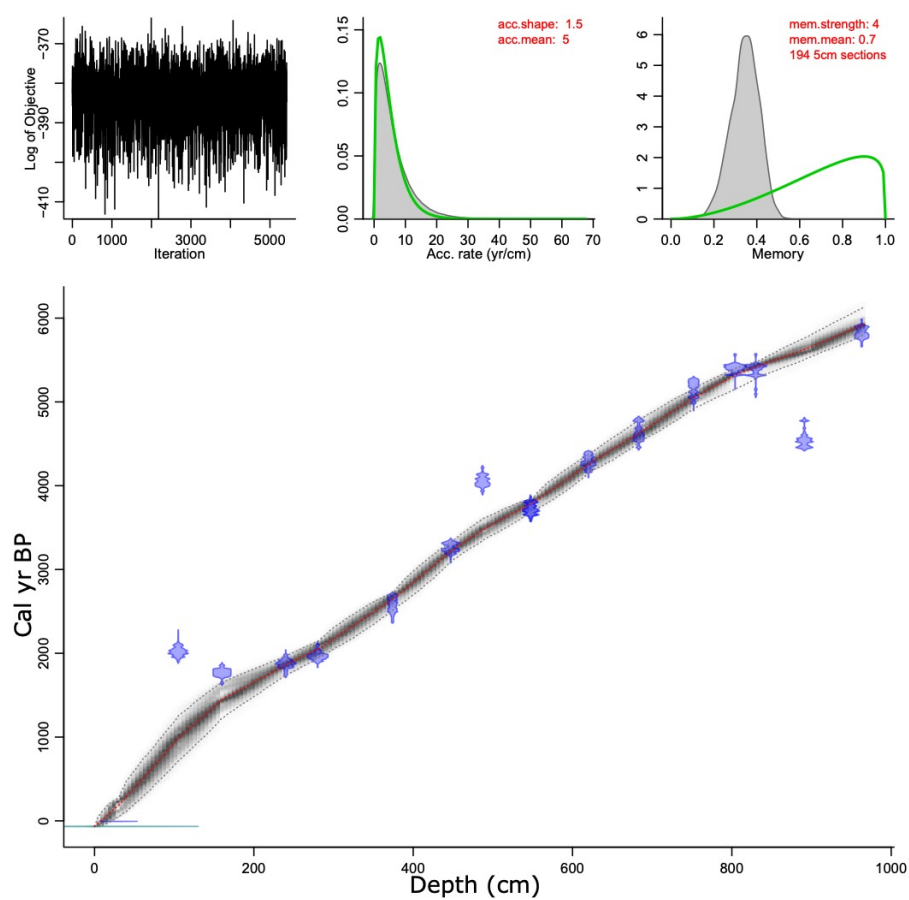


Figure 2. Age depth model at Lake Oltina



## References

- Akinyemi, F.O., Hutchinson, S.M., Mindrescu M, Rothwell. J.J.: Lake sediment records of atmospheric pollution in the Romanian Carpathians. *Quat Int* 293:105–113. doi:10.1016/j.quaint.2012. 01.022, 2013.
- Blaauw, M., and Christen, J.A.: Flexible paleoclimate age-depth models using an autoregressive gamma process. *Bayesian Analysis* 6, 457-474, <https://projecteuclid.org/euclid.ba/1339616472>, 2011.

- Begy, R.C., Kelemen, S., Simon, H., Tănăsolia, C.: The history of the sedimentation processes and heavy metal pollution in the Central Danube Delta (Romania). *Geochronometria*, 8: 10.1515/geochr-2015-0090 97-106, 2018.
- Hutchinson, S. M., Akinyemi, F. O., Mîndrescu, M., Begy, R., Feurdean, A.: Recent sediment accumulation rates in contrasting lakes in the Carpathians (Romania): impacts of shifts in socio-economic regime. *Reg Environ Change* **16**, 501–513. <https://doi.org/10.1007/s10113-015-0764-7>, 2016
- Reimer, P. J., Bard, E., Bayliss, A., Beck, J. W., Blackwell, P. G., Bronk Ramsey, C., Buck, C. E., Cheng, H., Edwards, R. L., Friedrich, M., Grootes, P. M., Guilderson, T. P., Haflidason, H., Hajdas, I., Hatté, C., Heaton, T. J., Hoffmann, D. L., Hogg, A. G., Hughen, K. A., Kaiser, K. F., Kromer, B., Manning, S. W., Niu, M., Reimer, R. W., Richards, D. A., Scott, E. M., Southon, J. R., Staff, R. A., Turney, C. S. M., van der Plicht, J.: IntCal13 and Marine13 radiocarbon age calibration curves 0-50,000 years cal BP, *Radiocarbon*, 55, 1869–1887, [https://doi.org/10.2458/azu\\_js\\_rc.55.16947](https://doi.org/10.2458/azu_js_rc.55.16947), 2013.
- Rose, N.L., Cogalniceanu, D., Appleby, P.G., Bancelj, A., Camarero, L., Fernandez, P.: Atmospheric contamination and ecological changes inferred from the sediment record of Lacul Negru in the Retezat National Park. *Advances in Limnology* 62, 319e350, 2009.