© *Acta hydrotechnica* **21/35 (2003)**, Ljubljana
ISSN 1581-0267

UDK/*UDC*: 519.61/.64:546.49:556.55
Kratki znanstveni prispevek – *Preliminary scientific paper*

Prejeto/*Received*: 7. 11. 2003
Sprejeto/*Accepted*: 30. 11. 2004

MODELIRANJE HIDRODINAMIKE IN TRANSPORTA ŽIVEGA SREBRA V VELENJSKEM JEZERU – 1. DEL: ŽIVO SREBRO V JEZERSKI VODI
MODELLING OF HYDRODYNAMICS AND MERCURY TRANSPORT IN LAKE VELENJE – PART I: MERCURY IN LAKE WATER

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Velenjsko jezero se nahaja v Šaleški dolini, v eni najbolj onesnaženih slovenskih regij. Večja vira onesnaženja sta dva: premogovna termoelektrarna v Šoštanju in Rudnik Velenje. Cilj te študije je bil ugotoviti transport in distribucijo živega srebra v Velenjskem jezeru. V preteklosti so bile posledice odlaganja premogovega pepela in odtočne vode z odlagališča pepela posebne kemične značilnosti jezerske vode, kot so na primer visoke pH vrednosti (10–12) in visoka vsebnost težkih kovin. Z uvedbo zaprtega kroga transportnega sistema pepela v letu 1995 se je kakovost vode hitro popravila. Da bi ugotovili novejše vire živega srebra v Velenjskem jezeru, smo izmerili koncentracije celokupnega živega srebra in metil-živega srebra na različnih okoljskih vzorcih (na dotoku v jezero, na iztoku, v deževnici, sedimentu itn.). Rezultati so pokazali, da so glavni vir živega srebra v jezeru jezerski pritoki in padavine. Koncentracije živega srebra in metil-živega srebra so v jezerski vodi izredno nizke (celokupno živo srebro: 0,2–2,7 ng/L; celokupno metil-živo srebro: 20–86 pg/L) in so primerljive z nekontaminiranimi jezeri po svetu. Koncentracije celokupnega in metiliranega živega srebra smo merili na površini in na različnih globinah, ugotavljali pa smo kroženje živega srebra, transport in kemične spremembe ter tudi za kalibracijo, preverjanje matematičnega modela za simulacije kroženja živega srebra v Velenjskem jezeru, kar bo podrobneje opisano v 2. delu prispevka. V 1. delu podajamo le kratek povzetek o izotopski sestavi jezerske vode ter vsebnosti Hg. Podatki o izotopski sestavi vode so bistvenega pomena za validacijo in verifikacijo modela, opisanega v drugem delu.

Ključne besede: *živo srebro, metil-živo srebro, matematično modeliranje, sladkovodno jezero*

Lake Velenje is located in one of the most polluted regions in Slovenia, the Šalek Valley. There are two major sources of pollution: the coal-fired thermal power plant in Šoštanj (ŠTPP) and the coal mine in Velenje. The aim of our study was to establish the transport and distribution of mercury in the Lake Velenje. In the past, dumping of coal ash directly into Lake Velenje and drainage water from the ash disposal site resulted in unique chemical characteristics of the lake water, such as very high pH (10–12) and high concentrations of heavy metals. The introduction of a closed cycle ash transport system in 1995 resulted in a very fast recovery of the lake water quality. With the aim of establishing recent sources of mercury in Lake Velenje, total mercury and methylmercury concentrations were measured in different environmental samples (lake inflows, outflow, rainwater, sediments, etc.). The results show that the major sources of mercury in Lake Velenje are lake inflows and wet deposition. Total mercury and methylmercury concentrations in the water column are very low (total mercury: 0.2–2.7 ng/L; methylmercury: 20–86 pg/L) and can be compared to other non-contaminated freshwater lakes. Total mercury and methylmercury concentrations were measured at the surface and at different depths to establish mercury cycling, its transport and chemical transformations, and for calibration and verification of a mathematical model for mercury cycling simulations in Lake Velenje, which is in detail described in Part 2. In Part 1 only short summary on lake water isotopic composition and Hg content is given. Data about isotopic composition of lake water are essential for validation and verification of the hydrodynamic model described in Part 2.

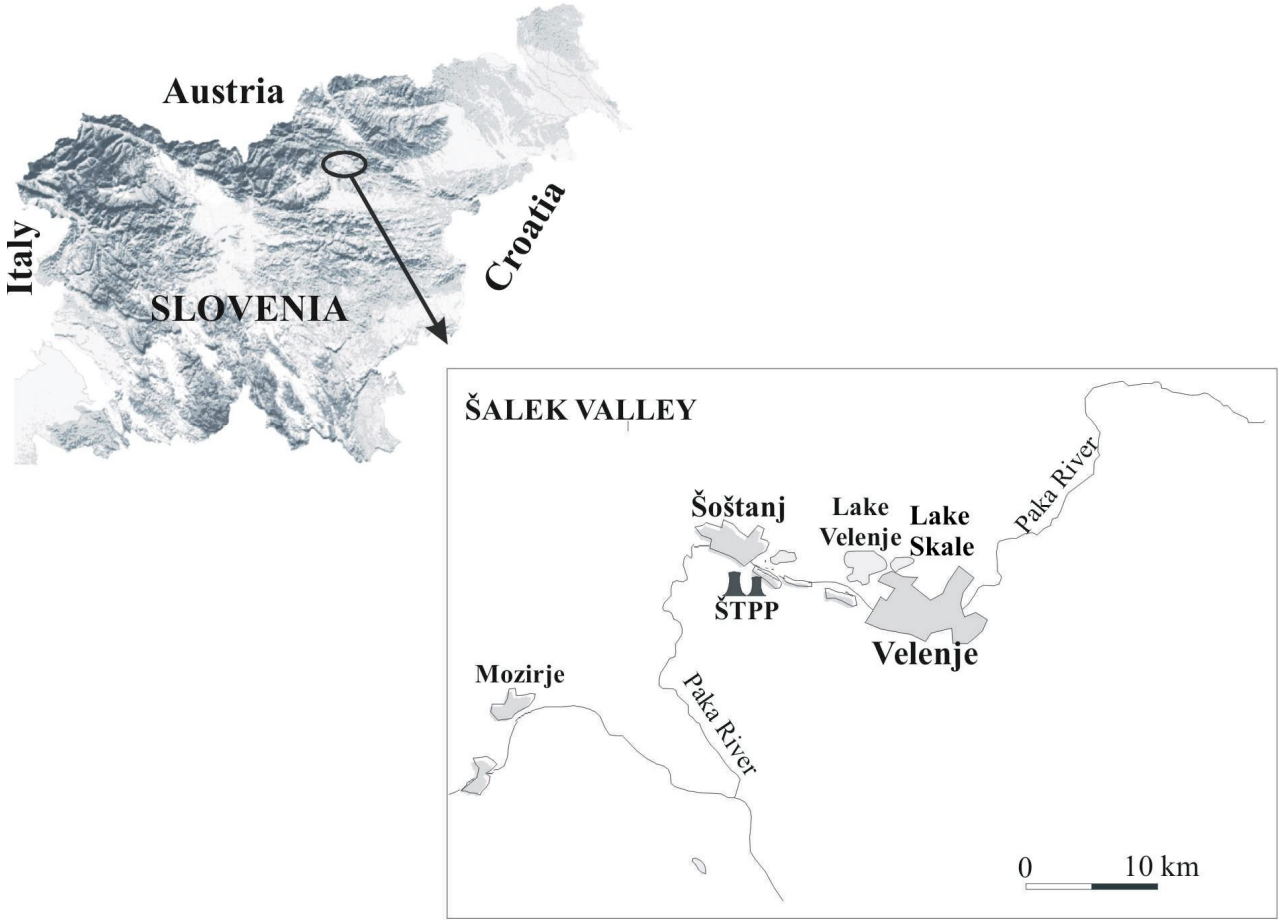
Key words: *mercury, methylmercury, mathematical modelling, freshwater lake*

1. UVOD

Onesnaženje zraka v bližini premogovnih termoelektrarn vpliva na celotni ekosistem ter tudi na človekovo zdravje. Sladke vode so pomemben del katerega koli ekosistema. Povišana koncentracija živega srebra v atmosferi pretežno vpliva na vodna telesa, in sicer neposredno z atmosferičnim usedanjem ali posredno z vtoki površinskih ali podzemnih voda. Tretjino slovenske elektrike proizvajajo premogovne termoelektrarne, med katerimi je Termoelektrarna Šoštanj (TEŠ) največja, s skupno proizvodnjo električne energije 775 MW (slika 1). V tem prispevku posvečamo pozornost porazdelitvi živega srebra v bližnjem Velenjskem jezeru. Območje jezera se namenja rekreativnim dejavnostim (ribarjenja, plavanja itn.). Za ugotavljanje vsebnosti in porazdelitve živega srebra ter kroženja Hg v Velenjskem jezeru smo uporabili dva pristopa.

1. INTRODUCTION

Air pollution in the vicinity of coal-fired thermal power plants affects the whole ecosystem as well as human health. Fresh waters are a very important part of any ecosystem. Elevated concentrations of mercury in the atmosphere mainly affect water bodies either directly through wet and dry deposition or indirectly by surface and ground water inflows. One third of Slovenian electricity is produced by coal-fired thermal power plants, among which the biggest one is the Šoštanj Thermal Power Plant (ŠTPP) with a common power of 775 MW (Figure 1). In this work attention was devoted to mercury distribution in nearby Lake Velenje. The lake is considered to be a recreational resource (e.g. fishing, swimming etc.). To establish mercury levels and its distribution in Lake Velenje and Hg cycling in the lake, two approaches were used.



Slika 1. Karta Slovenije s Šaleško dolino.
Figure 1. Map of Slovenia with the Šalek Valley.

Z monitoringom smo želeli ugotoviti nedavne vire in vsebnost živega srebra v okolju. Izmerjene podatke smo nato uporabili za modeliranje transporta živega srebra in njegovo porazdelitev v Velenjskem jezeru.

Hidrodinamični in transportni model smo uporabili iz več razlogov. Prvi razlog je bil, da je bilo zaradi visoke vrednosti pH jezero do 1995 biološko mrtvo. Po letu 1995 se je zaradi nove transportne tehnologije pepela jezerska voda zelo hitro popravila. Drugič, Velenjsko jezero je zaprt sistem, na katerega dotoki in iztoki ne vplivajo bistveno, kar je poenostavilo hidrodinamične izračune, modelno kalibracijo in preverjanje. Poleg tega je velika količina izmerjenih podatkov kakovosti jezerske vode in zraka v okolici jezera zmanjšala napake, ki bi jih v modelu povzročale aproksimacije. Dobro poznane so tudi meteorološke razmere v okolici jezera. In nazadnje, na območju je prisotno točkovno onesnaženje, ki je dobro poznano in predstavlja različne stopnje potencialne nevarnosti za okolje in človekovo zdravje, tako posredno kot neposredno.

Ker postopke vzorčenja, analitične metode in rezultate podrobneje opisujemo drugje (Kotnik *et al*., 1999; 2001), tukaj podajamo samo nekaj temeljnih rezultatov o vsebnosti živega srebra v jezeru in na dotokih v jezero (preglednica 1).

Through monitoring, we wished to establish recent sources and mercury levels in the environment. The measured data were then used further for modeling of mercury transport and distribution in Lake Velenje.

The application of a hydrodynamic and transport model to Lake Velenje was done for several reasons. First, until 1995, the lake was biologically dead, because of very high pH. After that year, ŠTPP's new ash transport technology resulted in a very fast recovery of the lake water quality. Second, Lake Velenje is a closed system with very little influence on water cycling from inflows and outflow, which simplified hydrodynamic calculations, model calibration and verification. In addition, lots of measured data of lake water quality and air quality in the surroundings of the lake reduced errors that would have been introduced into the model by approximations. Meteorological conditions in the vicinity of lake are also well known. Finally, there is a point source of pollution, which is well known and which represents a potential hazard to the environmental and human health, both directly and indirectly, through several mechanisms.

Since sampling procedures, analythical methods and results are in more detail described elsewhere (Kotnik *et al*., 1999; 2001) only some basic results about mercury content in the lake and its inflows are given in Table 1.

Preglednica 1. Koncentracija različnih vrst živega srebra na površini in v vodnem stolpu Velenjskega jezera – pomladne in zimske razmere (v ng/L).
Table 1. Concentration of different mercury species in surface and water column of Lake Velenje – spring and winter conditions (in ng/L).

	Mesto – *Location*	Celokupni Hg – *Total Hg*	Celokupni MeHg – *Total MeHg*
Pomlad – *Spring*	Povprečje v vodnem stolpu – *Average in water column* (Lake I; n = 16)	1.24	0.6197
	Dotok Lepene – *Lepena inflow* (n = 3)	2.68	0.0540
	Dotok Sopote – *Sopota inflow* (n = 3)	1.36	0.0680
	Lepena (n = 3)	4.34	0.1470
	Sopota (n = 3)	2.29	0.1010
	Iztok – *Outflow* (n = 3)	1.64	0.0390
Zima – *Winter*	Povprečje v vodnem stolpu – *Average in water column* (Lake I; n = 16)	0.97	0.025
	Dotok Lepene – *Lepena inflow* (n = 3)	2.48	0.015
	Dotok Sopote – *Sopota inflow* (n = 3)	1.90	0.030
	Lepena (n = 3)	2.39	0.066
	Sopota (n = 3)	1.00	0.005
	Iztok – *Outflow* (n = 3)	1.42	0.005

Velenjsko jezero ima dva dotoka. Potok Lepena priteka na vzhodni strani, poleg jeza, ki Velenjsko jezero deli od Škalskega jezera. Drugi dotok, potok Sopota, priteka v jezero s severa. Oba prispevata okoli 12.000.000 m^3 vode letno (ocena in meritve ERICO Velenje).

Oba potoka sta dokaj majhna. Potok Sopota izvira 4,5 km severno od pritoka v jezero. Teče skozi podeželsko območje, ki ga sestavljajo večinoma pašniki, travniki in gozdovi. Večji komunalni izpusti nanj ne vplivajo neposredno. Tok potoka je hiter v celi dolžini vodotoka. Povprečni letni pretok, izmerjen na dotoku v Velenjsko jezero, je 0,127 m^3/s (ocena in meritve ERICO Velenje).

Drugi dotok, potok Lepena, se nahaja na vzhodni strani jezera. Izvira okoli 4 km proti severovzhodu od vtoka v jezero, poleg manjšega naselja Cirkovce. Nato teče skozi Hrastovec. V povirnem delu na Lepeno vplivajo komunalni izpusti iz prej omenjenih naselij. Pred vstopom v Velenjsko jezero priteka v manjše Škalsko jezer, ki ga od Velenjskega jezera ločuje umetni jez iz sadre, odpadka procesa razžveplevanja zgorelega plina v TEŠ-u. Iz Škalskega jezera teče potok Lepena naprej v Velenjsko jezero. Med Škalskim in Velenjskim jezerom je ribogojnica, ki uporablja vodo iz Škalskega jezera. V ribogojnici gojijo predvsem postrvi in krape. Presežek vode iz ribogojnice se izliva v Lepeno okoli 100 m gorvodno od pritoka Lepene v Velenjsko jezero. Voda iz Velenjskega jezera teče v reko Pako in tako vpliva na kakovost rečne vode.

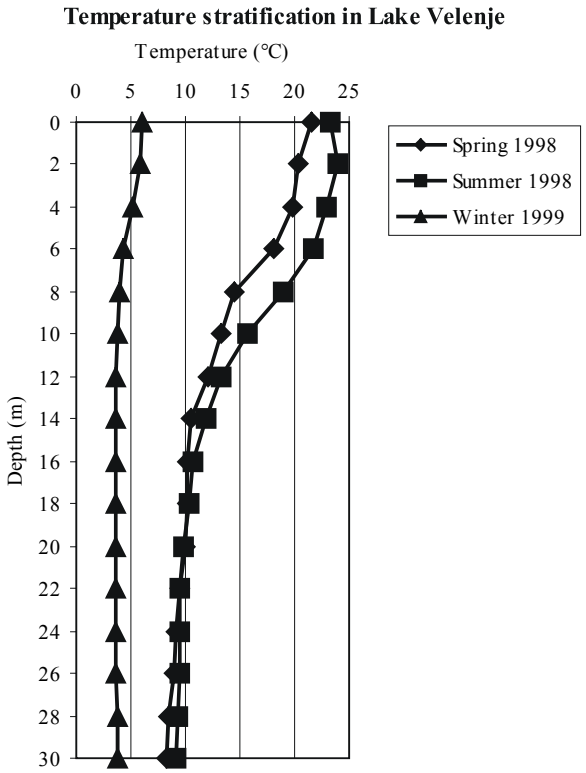
Velenjsko jezero ima tipične značilnosti majhnega sladkovodnega jezera z zmerno klimo. Ima močno termalno stratifikacijo med poletnimi in zgodnjimi jesenskimi meseci in je dobro mešano med zimskimi in zgodnjimi pomladnimi meseci (slika 2). Zgodaj spomladi je temperatura vode na vseh globinah približno 4 °C, zaradi povečanega sončnega sevanja se počasi začne segrevati. Segrete višje vodne plasti se zaradi vetra na površini nekaj časa mešajo z globljimi, hladnejšimi plastmi, s čimer se izenačujeta temperatura in z njo gostota vode po globini.

Velenje Lake has two inflows. The stream Lepena is on the eastern side near the dike that separates Lake Velenje from Lake Škale. The other, Stream Sopota, flows into the lake from the north. Both of them contribute 12,000,000 m^3 of water to the lake per year (measured and estimated by ERICO Velenje).

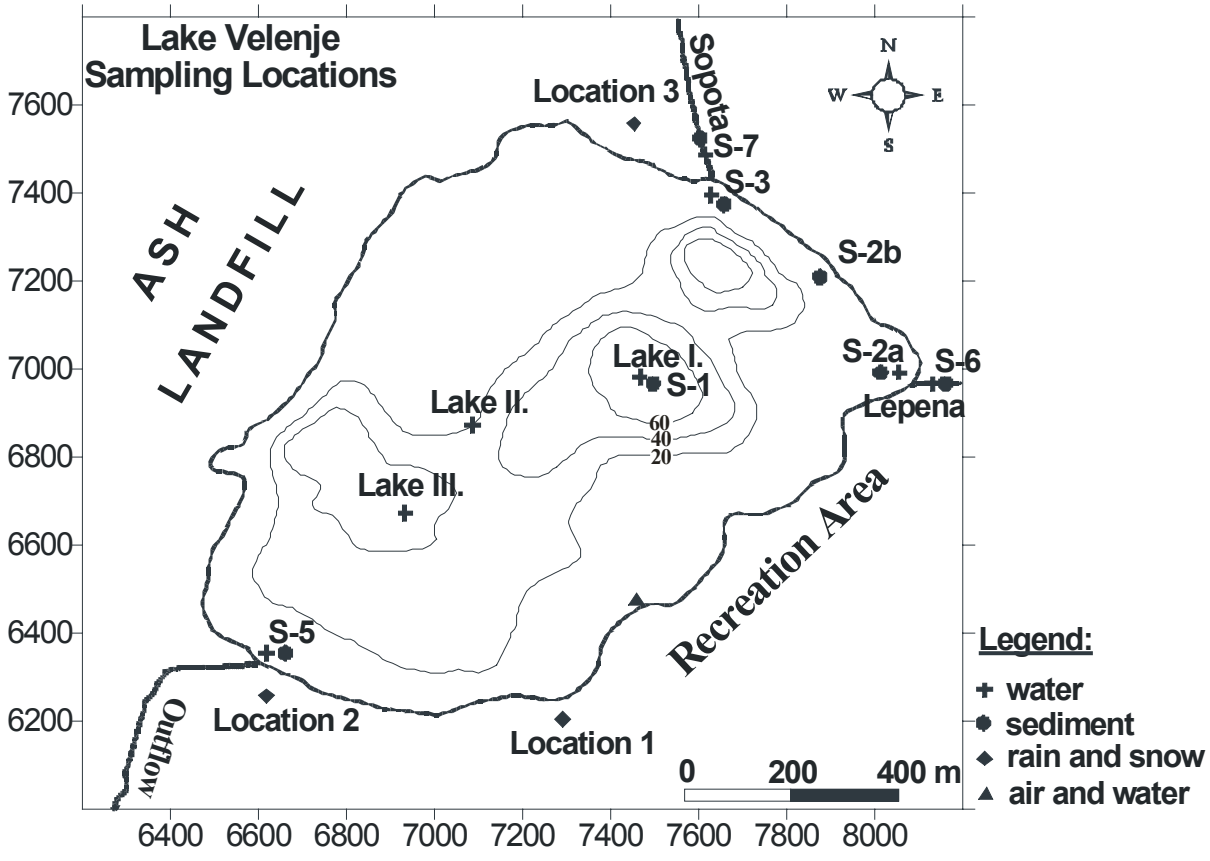
Both are relatively small streams. The Sopota stream has its source about 4.5 km north of its inflow into the lake. It runs through rural landscape, mostly consisting of pastures, meadows and woods. It is not directly influenced by larger municipal releases. The flow is quite fast throughout the whole Sopota bed. The average yearly flow measured at the inflow into Lake Velenje is 0.127 m^3/s (measured and estimated by ERICO Velenje).

A second inflow, the Lepena stream, is located at the eastern side of the lake. The Lepena stream has its source about 4 km to the northeast from its inflow into the lake, near a small settlement called Cirkovce. It runs through the settlement of Hrastovec. In its upper reaches Lepena stream is influenced by municipal releases from the aforementioned settlements. Before entering into Lake Velenje, it runs into the small Lake Škale that is separated from Lake Velenje by an artificial dyke made of gypsum, a waste product of the desulphurisation process of flue gases in ŠTPP. From Lake Škale, Lepena stream runs further into Lake Velenje. Between Lake Škale and Lake Velenje is a fish farm that uses water from Lake Škale. At the fish farm, mostly trout and carp are bred. Excess water from the fish farm is released into the Lepena stream about 100 m upstream from the Lepena inflow into Lake Velenje. Water from Lake Velenje flows into the Paka River and influences the water quality in that river.

Lake Velenje has typical characteristics of a small freshwater lake in a temperate climate. It has very strong thermal stratification during the summer and early fall months, and is well mixed during the winter and early spring months (Figure 2). In early spring, the water temperature at all depths is about 4°C and begins warming due to the increase of the solar radiation. The heated upper water layers mix with the deeper, colder layers until the density of the layers is equal.



Slika 2. Temperaturna stratifikacija Velenjskega jezera.
Figure 2. Temperature stratification in Lake Velenje.



Slika 3. Velenjsko jezero in odvzemna mesta.
Figure 3. Lake Velenje with sampling locations.

Ko pa je površinska plast (epilimnij; globina 0–4 m) dovolj segreta, tako da zaradi razlike v gostoti lahko plava nad hladnejšimi plastmi (hipolimnij, pod 16 m), se jezerska voda ne meša več. Navadno vetrovi nimajo dovolj energije, da bi vodo premešali do dna. Med dvema vodnima plastema je tranzicijska plast vode, kjer temperatura z globino hitro upada. Po temperaturni stratifikaciji v pozni pomladi veter meša le površinske plasti vode. Jezero se ohlaja podobno, kot se segreva: jeseni se površinska plast ohladi, medtem ko se temperatura epilimnija in hipolimnija izenači. Pri temperaturi 0 °C površinska plast zamrzne, s čimer se prepreči mešanje vode zaradi vetra.

When the surface layer (epilimnion; depth 0–4 m) is heated enough so that it can, because of density differences, float over the colder layers (hypolimnium; below 16 m) the water in the lake does not mix any more. Between the two water layers is a transitional water layer in which the temperature rapidly decreases with depth. After temperature stratification in the late spring, wind mixes only the surface water layer. The lake cools in the same way as it is heated: in fall, the surface layer is cooled, while temperatures between the epilimnion and hypolimnium become equal. At a temperature of 0°C the surface layer freezes, thus preventing the mixing of water due to winds.

2. VZORČEVANJE IN ANALIZE

Odvzemne točke okoli jezera in v jezeru so bile izbrane tako, da bi se zagotovilo detajlno predstavitev vsebnosti Hg, njegove transformacije ter transport v različnih okoljskih prostorih. Odvzemna mesta za jezersko vodo, vodo na dotoku, sedimente in padavine so prikazana na sliki 3.

Vzorci jezerske vode so bili odvzeti v treh sezonah: na koncu pomladi 1998 (začetek maja 1998), v sredini poletja 1998 (začetek avgusta 1998) in na koncu zime 1998/99 (konec februarja 1999). Vzorci so bili odvzeti na 8 mestih: 3 mesta so bila v sredini jezera, 3 na jezerskem dotoku in iztoku ter 2 na pritokih Lepene in Sopote. Na treh lokacijah v sredini jezera je vzorčenje potekalo na različnih globinah (0, 4, 8, 12, 16, 20, 24 in 30 m), ki so bile izbrane glede na stratifikacijo vode.

Raztopljeno celokupno živo srebro in metil-živo srebro, tako v raztopljeni obliki kot partikularno, sta bila merjena takoj po odvzemu. Celokupni THg je bil določen s tehniko CV AAS, in sicer po oksidaciji z BrCl, redukciji z $SnCl_2$ in ujetjem Hg^0 na zlato (Horvat *et al.*, 1996; Horvat *et al.*, 1991; Horvat, 1991). Reaktivno živo srebro v vodi je bilo določeno s tehniko CV AAS po redukciji s $SnCl_2$ in z amalgamacijo na zlato. Koncentracije MeHg v vodi so bile določene s tehniko CV AFS, in sicer po solventni

2. SAMPLING AND ANALYSES

The sampling points around and in Lake Velenje were chosen in order to provide a detailed representation of Hg content, its transformations and transport in different environmental compartments. Sampling locations for lake water, water inflow, sediment and precipitation are shown on Figure 3.

Lake water samples were taken in three seasons: at the end of spring 1998 (beginning of May 1998), in the middle of summer 1998 (beginning of August 1998) and at the end of winter 1998/99 (end of February 1999). Water samples were taken at 8 locations: three locations were in the centre of the lake, three at lake inflows and outflow points and two within the inflow streams of Lepena and Sopota. At the three locations in the middle of the lake, the samples were taken at different depths (0, 4, 8, 12, 16, 20, 24 and 30 m) that were chosen with regard to water stratification.

Dissolved and particulate phases of total and methylmercury in water samples were measured immediately after sampling. Determination of THg was performed by CV AAS following oxidation with BrCl, reduction with $SnCl_2$ and trapping of the resulting Hg^0 on gold (Horvat *et al.*, 1996; Horvat *et al.*, 1991; Horvat, 1991). Reactive mercury in water was determined by CV AAS following reduction by $SnCl_2$ and amalgamation on gold. MeHg concentrations in

ekstrakciji in vodni fazi etilacije MeHg (Horvat *et al.*, 1993a; 1993b; Liang *et al.*, 1994).

Vse analitične metode so se redno preverjale in izvajale v okviru kontrolnega sistema kakovosti Odseka za znanosti o okolju, Institut Jožef Stefan. Kakovost analitičnih meritev za analize Hg in speciacija sta se preverjali z redno uporabo certificiranih referečnih materialov (CRM), kot so DORM-1 (mišica trneža), DOLT-1 (jetra trneža) in IAEA-356 (onesnažen morski sediment). Po drugi strani pa za vodne vzorce certificirani referenčni materiali za celokupno, reakcijsko in metilirano živo srebro ne obstajajo, metode pa so bile vseskozi preverjane s protokolom US EPA (EPA Method 1631).

Izotopska sestava kisika v vodi je bila določena s standardno metodo, ki temelji na ravnovesju med referenčnim CO_2 pri 25 ^{o}C za 24 ur (Epstein in Mayeda, 1953), kar je bilo nato merjeno na masnem spektrometru Varian MAT 250. Uporabili smo notranje laboratorijske standarde (voda iz pipe – Ljubljanski vodovod z $\delta^{18}O$ = –9,34 ± 0,03 ‰ V-SMOW) za prepoznavanje morebitnih odstopanj med pripravo vzorcev ali instrumentalnih premikov med ugotavljanjem kisikovega izotopskega razmerja.

Postopek normalizacije za ^{18}O: normalizacijska enačba je bila pridobljena iz notranjih standardov, ki so bili merjeni v zadnjih treh mesecih po postopku programskega orodja za stabilne izotope LIMS v. 7.0.

Za določitev izotopske sestave devterija (^{2}H) je bila uporabljena redukcija vode na Cr pri 800 ^{o}C za proizvodnjo vodikovega plina (Gehre *et al*., 1996). Vodikov plin se nato izmeri na masnem spektrometru Varian MAT 250. Uporabljeni laboratorijski standardi so voda iz pipe in padavine: – 53,2 ± 1,4 ‰, – 32,4 ± 1,0 ‰, – 124,4 ± 1,2 ‰ V-SMOW. Pred analizo se laboratorijske standarde v serijah normalizira. Normalizacijsko enačbo smo pridobili po vsaj 25 meritvah po delovnih standardih, in sicer po postopku za stabilne izotope LIMS for Stable Isotopes v. 7.0.

water were determined by CV AFS following solvent extraction and aqueous phase ethylation of MeHg (Horvat *et al.*, 1993a; 1993b; Liang *et al.*, 1994).

All analytical methods were regularly validated and performed within a quality control system of the Department of Environmental Sciences, Jožef Stefan Institute. The quality of analytical measurements for Hg analysis and speciation was checked by the regular use of certified reference materials (CRMs), such as DORM-1 (Dogfish muscle), DOLT-1 (Dogfish liver) and IAEA-356 (Polluted marine sediment). As for water samples CRMs for total, reactive and methylmercury are not extant, methods were continuously validated by the US EPA protocol (EPA Method 1631).

The isotopic composition of oxygen in water was determined using the standard method based upon equilibration with referenced CO_2 at 25^{o}C for 24 h (Epstein and Mayeda, 1953), which was then measured on a Varian MAT 250 mass spectrometer. Internal laboratory standards were used (tap water – Ljubljanski vodovod with $\delta^{18}O$ = –9.34 ± 0.03 ‰ V-SMOW) to detect possible variations during sample preparation or instrumental drift during the oxygen isotopic ratio determination.

Normalization procedure for ^{18}O: the normalization equation was obtained from internal standards measured over the last 3 months following the procedure in the software LIMS for Stable Isotopes v. 7.0.

Reduction of water on Cr at 800^{o}C to produce hydrogen gas was used to determine the isotopic composition of deuterium (^{2}H) in water (Gehre *et al*., 1996). The hydrogen gas is then measured on a Varian MAT 250 mass spectrometer. The laboratory standards used are tap water and precipitation: – 53.2 ± 1.4 ‰, – 32.4 ± 1.0 ‰, – 124.4 ± 1.2 ‰ V-SMOW. The laboratory standards are normalized in batches before the samples are analyzed. The normalization equation was obtained from at least 25 measurements of working standards following the procedure in the software LIMS for Stable Isotopes v. 7.0.

The calibration of the $\delta^{18}O$ and $\delta^{2}H$ values analyzed with respect to the international

Kalibracija vrednosti $\delta^{18}O$ in $\delta^{2}H$, analizirana v skladu z mednarodnimi standardi, je bila izvedena z analizo VSMOW, GISP in SLAP (‰ vrednosti) z istimi postopki. Datum zadnje kalibracije je januar 1999.

Rezultati $\delta^{18}O$ in $\delta^{2}H$ so prikazani kot deviacije v ‰ od standarda V-SMOW (Coplen, 1994). Natančnost analize $\delta^{18}O$ in $\delta^{2}H$, ki je temljila na ponovljenih meritvah, je bila ± 0,05 ‰ oziroma ± 1,0 ‰.

3. Hg V JEZERSKI VODI IN SEDIMENTIH

Koncentracije celokupnega Hg (THg) in celokupnega MeHg (TMeHg) (preglednica 1, sliki 4 in 5) v Velenjskem jezeru so zelo nizke v primerjavi z drugimi onesnaženimi jezeri severne poloble (Kim in Fitzgerald, 1986; Henry *et al*., 1995b). Koncentracije Hg in MeHg v površinskem sloju in vodnem stolpu so primerljive s koncentracijami, ki so bile izmerjene v neonesnaženih, oddaljenih sladkovodnih jezerih severne poloble (Meuleman *et al*., 1995; Henry *et al.,* 1995b; Leermakers *et al*., 1996). Delež metiliranega živega srebra MeHg je 2,01–24,41 %, kar lahko primerjamo s podatki iz literature, ki za sladkovodne sisteme navaja 1–12 % (Lee & Iverfeldt, 1991) in 7–25 % delež (Bloom et al. 1991).

Pri vzorcih, odvzetih spomladi, so bile najvišje koncentracije celokupnega THg ugotovljene pri površinski jezerski vodi pri dotoku Lepene (2,68 ng/L), to pa smo pripisali visoki vsebnosti partikularnega Hg (75,5–81,8 %) v potoku Lepena. Koncentracije THg v jezeru so se manjšale od vzhoda (dotok Lepene) proti zahodu. Najvišje vrednosti THg v vodnem stolpu so bile v površinskem sloju (globine O–4 m). Pod globino 4 m so se koncentracije THg zmanjšale na vrednost 1 ng/L. Večina živega srebra v vodnem stolpu je bila vezana na (trdne) delce (50–60 %). Spomladi so bile najvišje koncentracije TMeHg ugotovljene na dotokih Lepene (54 pg/L) in Sopote (68 pg/L). Povečano koncentracijo MeHg proti jezerskemu dnu lahko pripišemo metilaciji Hg v sedimentu in difuziji MeHg, ki prihaja iz sedimenta v vodo nad njim (Sukhenko in Vasiliev, 1995).

standards was carried out by analyzing VSMOW, GISP and SLAP (‰ values) with the same procedures. The date of the last calibration is January 1999.

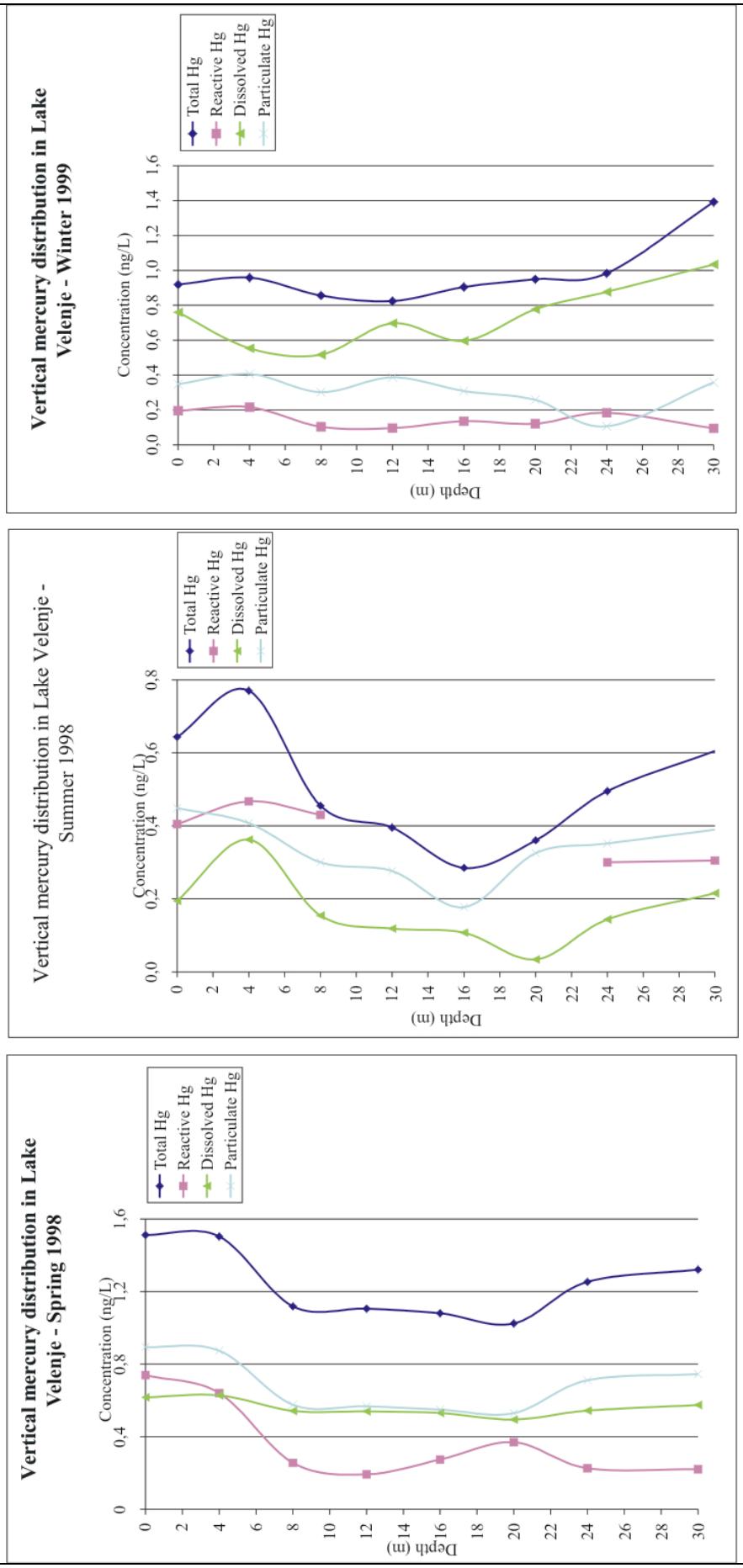
$\delta^{18}O$ and $\delta^{2}H$ results are reported as deviations in ‰ from the V-SMOW standard (Coplen, 1994). The precision of the analyses of $\delta^{18}O$ and $\delta^{2}H$ based upon replicate measurements was ± 0.05 ‰, and ± 1.0 ‰, respectively.

3. Hg IN LAKE WATER AND SEDIMENTS

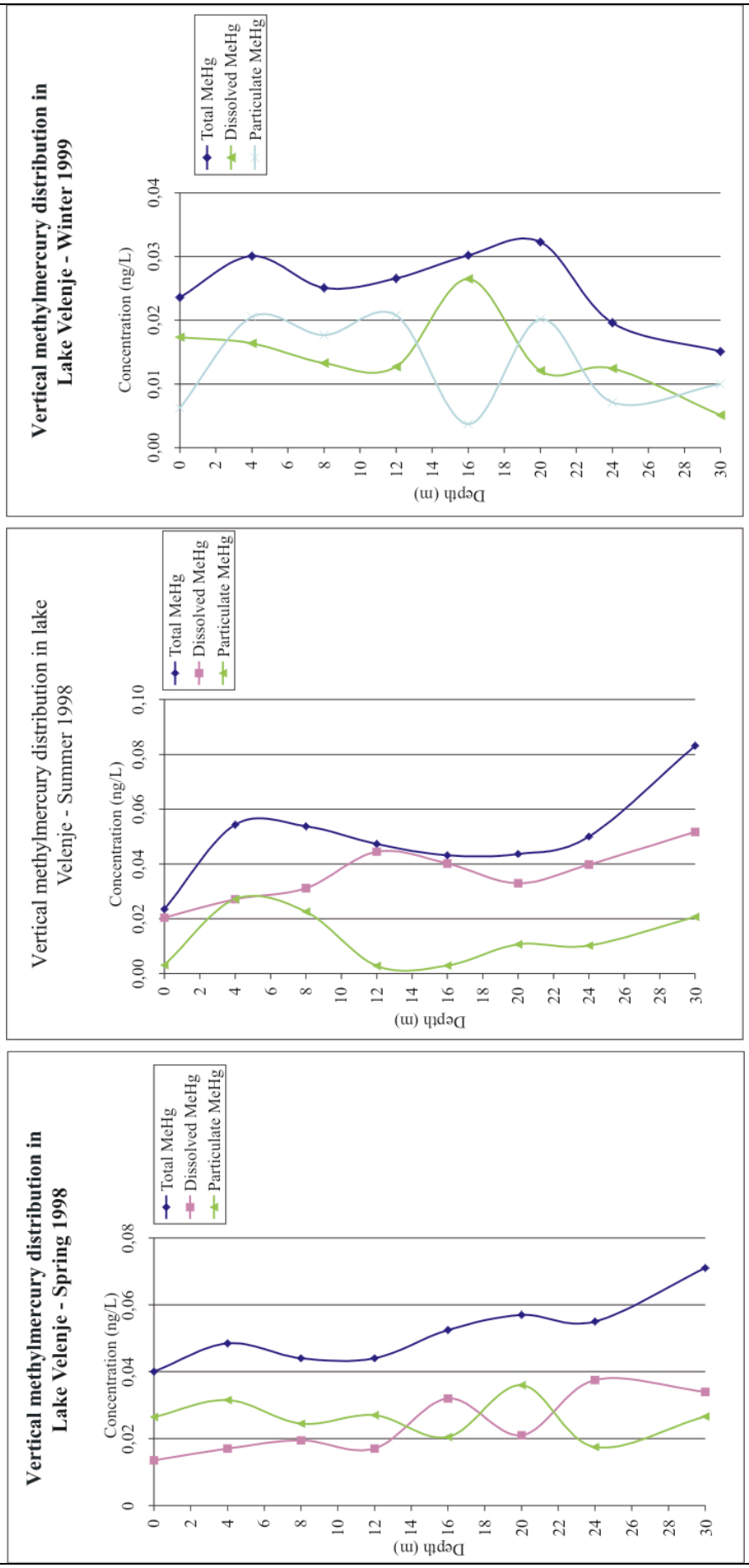
Total Hg (THg) and total MeHg (TMeHg) concentrations (Table 1, Figures 4 and 5) in Lake Velenje are very low compared to Hg concentrations in other polluted lakes of the northern hemisphere (Kim and Fitzgerald, 1986; Henry *et al*., 1995b). Hg and MeHg concentrations in the surface layer and water column are comparable to those reported for unpolluted remote freshwater lakes of the northern hemisphere (Meuleman *et al*., 1995; Henry *et al.,* 1995b; Leermakers *et al*., 1996). The percentages of MeHg are between 2.01 % and 24.41 % and are comparable to the literature data, which report 1–12 % (Lee & Iverfeldt, 1991) and 7–25 % (Bloom et al. 1991) for freshwater systems.

Of the samples taken during spring, the highest THg concentrations were found in the lake surface water at the Lepena inflow (2.68 ng/L) due to the high content of particulate Hg (75.5–81.8 %) in Lepena Stream. THg concentrations in the lake decreased from east (Lepena inflow) to west. The highest values of THg in the water column were in the surface layer (0–4 m of depth). Below 4 m of depth, the THg concentrations decreased to a value of 1 ng/L. Most mercury in the water column was bounded to particulate matter (50–60 %).

The highest TMeHg concentrations during spring were found in the Lepena (54 pg/L) and Sopota (68 pg/L) inflows. The increase in MeHg concentrations towards the lake bottom is due to the methylation of Hg in sediment, and the diffusion of the MeHg produced from the sediment into the overlaying water (Sukhenko and Vasiliev, 1995).



Slika 4. Vertikalna razporeditev različnih oblik živega srebra v Velenjskem jezeru v različnih letnih časih.
Figure 4. Vertical distribution of different mercury species in Lake Velenje during diferrent seasons.



Slika 5. Vertikalna razporeditev različnih oblik metil živega srebra v Velenjskem jezeru v različnih letnih časih.
Figure 5. Vertical distribution of different methylmercury species in Lake Velenje during diferrent seasons.

Poletne koncentracije THg v jezeru so bile nižje kot pomladne (0,64–0,77 ng/L). V vseh primerih je bila najnižja vrednost THg v mezolimniju (0,03–0,12 ng/L). Odstotni delež partikularnega Hg v vodnem stolpu je bil med 50 in 80 %. Koncentracije celokupnega MeHg v površinskem sloju so bile najvišje pri dotoku Lepene in so se znižale proti zahodnemu delu jezera. Podobno stanje smo ugotovili pri partikularnem MeHg. Raztopljeno MeHg je bilo enakomerno porazdeljeno po jezerski površini. Intenzivnejše koncentracije MeHg poleti v primerjavi s pomladnimi so posledica višjih letnih temperatur vode, s tem pa intenzivnejših makrobiotskih aktivnosti ter procesov metilacije, demetilacije in redukcije v samem jezeru in pritokih.

Pozimi je bila najvišja koncentracija THg izmerjena na dotoku Lepene (2,48 ng/L). Večina živega srebra na tem mestu je bila vezana na delce (75 %). V površinskem sloju koncentracije Thg naraščajo od zahodnega proti vzhodnemu delu jezera. V vodnem stolpu je vrednost THg naraščala z globino, in sicer od 0,9 ng/L na 1,4 ng/L. Najvišje koncentracije TMeHg smo našli na dotoku Lepene, in sicer v razponu med 15 in 35 pg/L. Razmerje med partikularnimi in raztopljenimi oblikami MeHg je bilo skoraj enako v celotnem vodnem stolpu (0,4 do 2,7 %).

Celokupna koncentracija živega srebra je v dotoku Lepene za spoznanje višja kot v jezeru ali pri potoku Sopota (glej preglednico 1). Višje koncentracije Hg in MeHg so lahko posledica značilnosti prispevnega območja potoka Lepena. Lepena teče skozi več manjših vasi, ki svoje komunalne odplake spuščajo neposredno v potok. Preden se Lepena izteče v Velenjsko jezero, ta oblikuje manjše jezero (Škalsko jezero). Voda iz Škalskega jezera se uporablja za ribogojstvo in se nato vrača v Lepeno. Višje koncentracije THg v Lepeni so verjetno posledica komunalnih odplak in ribogojstva (i. e. fungicidi in pesticidi z vsebnostjo Hg) v vaseh na povodju Lepene. Koncentracija MeHg v potoku Lepena pa je posledica biotske metilacije Hg v Škalskem jezeru in dodatne biotske metilacije v ribogojnici.

Najbolj informativni element vodnega sistema za presojanje ravni stalne kontaminacije so talni sedimenti. Temeljni sediment v

The summer THg concentrations in the lake were lower than the spring concentrations (in a range between 0.64 and 0.77 ng/L). In all cases the lowest THg content was in the mesolimnion (between 0.03 and 0.12 ng/L). The percentage of particulate Hg in the water column was between 50 and 80 %. The total MeHg concentrations in the surface layer were highest at the Lepena infow, and decreased toward the western part of the lake. A similar situation was also observed for particulate MeHg. Dissolved MeHg was uniformly distributed across the lake surface. The more intense MeHg concentration peaks in the summer compared to the spring are caused by the higher water temperatures in the summer, and with that more intense microbial activity, methylation, demethylation and reduction processes in the lake and its inflows.

In winter the highest THg content was found at the Lepena inflow (2.48 ng/L). Most mercury at that location was bound to particulate matter (75 %). In the surface layer, THg concentrations increased from the western part to the eastern part of the lake. In the water column, THg content increased with depth from 0.9 to 1.4 ng/L. The highest TMeHg concentrations were found at the Lepena inflow. TmeHg concentrations were in the range between 15 and 35 pg/L. The proportion between particulate and dissolved MeHg forms was almost the same all throughout the water column (0.4 to 2.7 %).

Among inflows total mercury concentrations in the Lepena Stream are slightly higher than in the lake or in Sopota Stream (see Table 1). The higher concentrations of Hg and MeHg in the Lepena Stream can be explained by the characteristics of the Lepena Stream catchment area. Lepena Stream flows through few small villages that release their municipal discharges directly into the stream. Before the Lepena Stream enters Lake Velenje, it forms a small lake (Lake Škale). Water from Lake Škale is further used for fish-farming and returned back to the Lepena Stream. Higher THg concentrations in the Lepena are probably a consequence of municipal waste discharges and farming activities (i.e. fungicides and pesticides containing Hg) in the villages of its catchment area. MeHg in the Lepena Stream has its origin in biotic methylation of Hg in Lake Škale and additional biotic methylation at the fish farm.

The most informative component of water

Velenjskem jezeru je premogov pepel, ki se je v preteklosti odlagal v jezeru. V tistih delih jezera, ki so zaprti za dotoke, sedimenti vsebujejo več organskih snovi. Koncentracije THg v vzorcih sedimenta iz odvzemnih mest v Velenjskem jezeru so bile spremenljivih vrednosti, od 53 do 166 ng/g (suhe teže). Višje koncentracije THg so bile ugotovljene v organsko bogatem sedimentu v bližini dotokov v jezero. V globljih delih jezera, kjer sediment večinoma tvori premogov pepel, so bile koncentracije THg dokaj nizke 71,5 ng/g, suhe teže). Tudi koncentracije MeHg so bile višje v organsko obogatenem sedimentu v bližini dotokov v jezero kot v premogovem pepelu, ki se je odlagal v globljih delih jezera. Najnižja koncentracija MeHg je bila ugotovljena na najnižji točki jezera (0,05 ng/g, suhe teže). Povišani koncentraciji THg in MeHg ob obeh dotokih sta posledica sedimentacija organsko obogatenih sedimentov iz potokov Lepene in Sopote.

4. δ^{18}O IN δ^{2}H

Izotopi so atomi, katerih jedra vsebujejo enako število protonov, a različno število nevtronov. Izotopi istega elementa imajo različne fizikalno-kemične lastnosti. Delimo jih v dve temeljni skupini: na stabilne in nestabilne (radioktivne) izotope.

Izotopsko sestavo ali razmerje med težjimi in lažjimi izotopi istega elementa v spojini izražamo z vrednostjo δ, ki je relativna razlika med izotopsko sestavo vzorca in standardom. Vrednost δ ponavadi izražamo v tisočinkah ($^0/_{00}$):

systems, from the standpoint of estimating their degree of stable contamination, is bottom sediments. The sediment in Lake Velenje is mostly coal ash deposited into the lake in the past. In the parts of the lake close to lake inflows, the sediment contains more organic matter. The THg concentrations in sediment samples from locations in Lake Velenje varied between 53 and 166 ng/g (dry weight). Higher THg concentrations were found in organic-rich sediment close to the lake inflows. In the deeper parts of the lake THg concentrations in the sediment, which consisted mostly of coal ash, the concentrations were relatively low (71.5 ng/g, dry weight). MeHg concentrations were also higher in the organic-rich sediment close to the lake inflows than in the coal ash that is deposited in the deeper parts of the lake. The lowest MeHg concentration was found in the deepest point of the lake (0.05 ng/g, dry weight). Increased THg and MeHg concentrations near both inflows are a consequence of sedimentation of organic-rich sediment from the Lepena and Sopota streams.

4. δ^{18}O AND δ^{2}H

Isotopes are atoms whose nuclei contain the same number of protons but different numbers of neutrons. Isotopes of the same element have different physico-chemical properties. Isotopes can be divided into two fundamental kinds: stable and unstable (radioactive).

The isotopic composition, or proportion between heavier and lighter isotopes of the same element in a compound, expressed by the value δ, which is the relative difference between the isotopic composition of a sample and a standard. Usually the δ value is expressed in per mill ($^0/_{00}$):

$$\delta A = \frac{R_{sample} - R_{standard}}{R_{standard}} \cdot 1000\,, \qquad (1)$$

kjer je A težji izotop in $R = [A_2]/[A_1]$, je razmerje med koncentracijo lažjega $[A_1]$ in težjega $[A_2]$ izotopa v spojini vzorca ali standarda.

Delitev izotopov na dve snovi ali dve fazi iste snovi z različnimi izotopskimi razmerji imenujemo "izotopsko frakcioniranje". Temeljni dejavniki, ki povzročajo izotopsko

where A is the heavier isotope and $R = [A_2]/[A_1]$ is the proportion between the concentration of the lighter $[A_1]$ and heavier $[A_2]$ isotope in a compound of sample or standard.

The partitioning of isotopes between two substances or two phases of the same substance with different isotope ratios is called

frakcioniranje, so izotopske reakcije izmenjave in kinetični procesi, ki temeljijo predvsem na razlikah v reakcijah, stopnjah izotopskih molekul, ki jih povzročajo razlike v masi, ter na temperaturi.

S faktorjem frakcioniranja (α) merimo stopnjo izotopske frakcionacije med spojinami in ga definiramo, kot sledi:

"isotope fractionation". The main phenomena producing isotope fractionation are isotope exchange reactions and kinetic processes, which depend primarily upon differences in the reactions, rates of isotopic molecules caused by their mass differences, and upon temperature.

The fractionation factor is a measure of the degree of isotope fractionation between compounds, and is defined as follows:

$$\alpha_{A-B} = \frac{R_A}{R_B} = \frac{1000 + \delta A}{1000 + \delta B}, \tag{2}$$

kjer je *R* razmerje koncentracije med težjim in lažjim izotopom v spojini *A* in spojini *B*.

Poznavanje izotopske sestave vode nam lahko pomaga pri ugotavljanju vira in sledenju njegovih poti. Ko voda na primer izhlapeva s površine morja ali jezer, se vodna para obogati v lažjih izotopih H in ^{16}O, saj ima $H_2{}^{16}O$ višji parni/uparjeni pritisk kot H^2HO in $H_2{}^{18}O$. Med odstranitvijo dežja iz vlažnih zračnih mas se ostanek vlage trajno siromaši v težkih izotopih, saj je dež, ki zapušča sistem, obogaten z ^{18}O and ^2H. Če se zračne mase premikajo proti polu in se ohlajajo, bo dodatni dež, ki se tvori, vseboval manj ^{18}O kot začetni dež. V zmernih in vlažnih podnebjih je izotopska sestava podtalnice in tekočih površinskih voda podobna sestavi padavin v območju obogatitve (Gat, 1971). Ko voda vstopi v jezero ali drugo območje izhlapevanja, lahko pričakujemo, da se bo zelo obogatila s težkima izotopoma ^2H in ^{18}O.

Rezultati meritev stabilnih izotopov $\delta^{18}O$ in $\delta\ ^2H$ v Velenjskem jezeru so prikazani v preglednici 2.

Na kisikovo izotopsko sestavo v vodnem stolpu lahko vpliva več dejavnikov. V naravnih pogojih, predvsem pa pri jezerih srednje zemljepisne širine, vrednost $\delta^{18}O$ jezerske vode nikoli ne ostane konstanta. Nasprotno, vrednost niha okoli vrednosti stacionarnega stanja/stanja mirovanja, ustrezujoč časovnim spremembam različnih dejavnikov (temperature, relativne vlažnosti, izotopske sestave skupnega dotoka in atmosferične vodne pare).

where *R* is concentration proportion between the heavier and lighter isotope in compound *A* and compound *B*.

Knowledge of water isotopic composition can help us to determine its source and trace its pathways. For instance when water evaporates from the surface of the ocean or from lakes, the water vapor is enriched in lighter isotopes H and ^{16}O because $H_2{}^{16}O$ has a higher vapor pressure than H^2HO and $H_2{}^{18}O$. During removal of rain from moist air mass, the residual vapor is continuously depleted in the heavy isotopes, because the rain leaving the system is enriched in ^{18}O and ^2H. If the air mass moves poleward and becomes cooler, the additional rain that is formed will contain less ^{18}O than the initial rain. In temperate and humid climates the isotopic composition of groundwater and flowing surface water is similar to that of the precipitation in the area of recharge (Gat, 1971). When water enters a lake or other evaporative environment, it can be expected to undergo extreme enrichments in the heavy isotopes ^2H and ^{18}O.

The results of the $\delta^{18}O$ and $\delta\ ^2H$ stable isotope measurements in Lake Velenje are shown in Table 2.

There are several factors that can affect the oxygen isotopic composition in the water column. Under natural conditions, particularly for mid-latitude lakes, the $\delta^{18}O$ value of lake waters never remains constant. Instead, it fluctuates around the steady-state value, responding to seasonal changes of respective parameters (temperature, relative humidity, isotopic composition of total inflow and atmospheric water vapour).

Preglednica 2. $\delta^{18}O$ in $\delta\ ^{2}H$ v Velenjskem jezeru in na dotokih (v $^{o}/_{oo}$).
Table 2. $\delta^{18}O$ and $\delta^{2}H$ in Lake Velenje and its inflows (in $^{o}/_{oo}$).

Mesto/*Location*	**Globina (m)/*Depth (m)***	**Pomlad/*Spring***		**Zima/*Winter***	
		$\delta^{18}O$ [‰]	$\delta^{2}H$ [‰]	$\delta^{18}O$ [‰]	$\delta^{2}H$ [‰]
Jezero I/*Lake I*	0	- 7,09	- 55,5	- 7,92	- 59,2
	4	- 7,21	- 56,5	- 7,97	- 58,7
	8	- 7,49	- 56,7	- 7,81	- 58,0
	12	- 7,66	- 57,2	- 7,74	- 57,7
	16	- 7,57	- 56,9	- 7,19	- 56,8
	20	- 7,56	- 57,9	- 7,80	- 58,8
	24	- 7,38	- 58,2	- 7,86	- 57,6
	30	- 7,34	- 57,7		
Jezero II/*Lake II*	0			- 7,83	- 57,2
	4			- 7,84	- 56,8
	8			- 7,83	- 56,6
	12			- 7,89	- 57,3
	16			- 7,78	- 56,8
	20			- 7,82	- 56,6
	24			- 7,77	- 57,3
Jezero III/*Lake III*,	0	- 7,05	- 54,6	- 6,94	- 57,4
	4	- 6,88	- 52,9	- 7,96	- 56,9
	8	- 7,46	- 55,8	- 7,88	- 57,2
	12	- 6,93	- 57,6	- 7,78	- 58,6
	16	- 7,13	- 56,9	- 7,82	- 57,9
	20	- 7,43	- 55,9	- 7,80	- 57,2
	24	- 7,49	- 56,0		
	30	- 7,60	- 57,7		
Dotok Lepene/*Lepena inflow*	0	- 7,34	- 57,8	- 8,95	- 62,9
Dotok Sopote/*Sopota inflow*	0	- 6,85	- 54,2	- 9,49	- 66,4
Odtok/*Outflow*	0	- 6,98	- 53,9	- 7,86	- 57,7
Lepena	0	- 8,98	- 62,1	- 9,53	- 65,2
Sopota	0	- 8,57	- 59,5	- 9,47	- 65,2

Amplituda sprememb zaradi letnih časov je odvisna od specifičnih podnebnih razmer določenega jezera, vendar je ponavadi nekje med 1 in 2 ‰. V sezonsko stratificiranih jezerih se aktivni volumen jezera spreminja z letnim časom. Med poletno stratifikacijo je izotopsko bogatenje jezera zaradi povečanega izhlapevanja omejeno na zgornjo, dobro premešano plast (epilimnij). Zimski preobrat pa zmanjša vrednost $\delta^{18}O$ zgornjega sloja zaradi mešanja z izotopsko lažjimi vodami hipolimnija.

The amplitude of these seasonal variations depends upon the particular climatic setting of the given lake but is usually in the range of 1 to 2 ‰. In seasonally stratified lakes the active volume of the lake changes with the season. During the summer stratification, isotopic enrichment of the lake water due to enhanced evaporation is limited to the upper, well-mixed layer (epilimnion). Winter turnover reduces the $\delta^{18}O$ value of the upper layer due to mixing with isotopically lighter hypolimnion waters.

5. ZAKLJUČKI

V splošnem smo višje koncentracije THg v vodnem stolpu našli v epilimniju, in sicer zaradi vnosa raztopljenega in partikularnega Hg na dotokih. V bližini odlagališča nismo izmerili povečanih koncentracij THg in MeHg. Koncentracije THg in TMeHg v Velenjskem jezeru so bistveno nižje od koncentracij Hg v drugih onesnaženih jezerih severne poloble. Velenjsko jezero ne predstavlja potencialnega vira Hg in MeHg v človekovi prehranjevalni verigi, četudi bi se jezero spremenilo v visoko bioproduktivno jezero.

V drugem delu prispevka bodo predstavljeni podatki o izotopski sestavi in vsebnosti različnih zvrsti živega srebra v pritokih in jezerski vodi, uporabljeni za validacijo in verifikacijo hidrodinamskega in geokemičnega modela kroženja in pretvorb živega srebra in njegovih spojin v Velenjskem jezeru. Končni cilj raziskave so simulacije različnih možnih scenarijev spreminjanja vsebnosti Hg v jezeru.

5. CONCLUSIONS

Generally, higher concentrations of THg in the water column were found in the epilimnion, due to the input of dissolved and particulate Hg from inflows. In the vicinity of the landfill we did not find any elevated THg or MeHg concentrations. THg and TMeHg concentrations in Lake Velenje are very low compared to Hg concentrations in other polluted lakes of the northern hemisphere. Lake Velenje does not represent a potential source of Hg and MeHg in human food chain even in case that it turns onto high bioproductive lake.

Presented data on isotopic composition and different Hg species in inflows and lake water were further used for validation and verification of the hydrodinamical and geochemical model of Hg cycling and its transformations in Lake Velenje that is represented in second part. The final aim of the study are simulations of Hg behaviour under different possible scenarios.

ZAHVALA

Za finančno podporo se zahvaljujemo Ministrstvu za šolstvo, znanost in šport Republike Slovenije.

ACKNOWLEDGEMENTS

We acknowledge the financial support of the Ministry of Education, Science and Sports of Slovenia for funding this work.

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