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# Estimation of mercury mass balance in the Gulf of Trieste

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*The annual mercury mass balance of the Gulf of Trieste, which includes inflows from the river Soča and from the open sea of the Northern Adriatic, (wet) atmospheric deposition and bottom sediment resuspension is presented. The sinks are the outflow to the open sea and burial. The river Soča inflow (1500 kg Hg year$^{-1}$, approx.) and bottom burial (1300 kg Hg year$^{-1}$, approx.) are the two major items of the mass balance. In spite of some strong resuspension, 82 % of mercury is removed from the water column by burial. On the other hand, the bottom sediment is a major source of monomethyl mercury (MMHg) for the Gulf, with a net upward flux of 90 kg of MMHg year$^{-1}$ at the sediment-water interface.*

**Key words:** marine pollution, mercury, mass-balance, the river Soča, Gulf of Trieste

## INTRODUCTION

Recent studies have shown that the former mercury mining area around the town of Idrija, Slovenia, continues to supply high quantities of mercury to the river Idrijca, although mining and smelting activities ceased some ten years ago (e.g. GOSAR *et al.*, 1997a, 1997b; HORVAT *et al.*, in press; HINES *et al.*, in press). The reason for persisting pollution is erosion of and direct runoff from the tailings and other contaminated soils along the river. The 30 km long river Idrijca is a tributary of the river Soča, which flows for a further 70 km before discharging into the Gulf of Trieste, the NE part of the Adriatic Sea (Fig. 1). Due to the transport effect of this river system, mercury pollution occurs in areas far away from the primary source. Moreover, 10 years after shutdown of mercury production and 4 years after the complete closure of the Idrija mine in 1995, no appreciable decrease of mercury concentrations was observed in this marine environment (HORVAT *et al.*, 1996, 1998; COVELLI *et al.*, 1999a).

In 1996, a provisional annual mercury mass balance was established for the Gulf of Trieste (ŠIRCA, 1996; ŠIRCA *et al.*, 1999). Some


*Fig. 1. Situation of the Gulf of Trieste and of the mainland area considered*

recent measurements of mercury concentrations at the bottom of the Gulf, in the river Soča water and in particulate matter enabled a revision and adaptation of the mass balance, especially for monomethyl mercury (MMHg) data. This mass balance is especially important in the initial stage of development of a 3D model of mercury transport and fate in the Gulf (ŽAGAR *et al.*, 1999) in which the main processes and mercury transformations have to be defined.

## THE RIVER SOČA

### Hydrology

Hydrology of the alpine river Soča is well known in the cross-section at Solkan in Slovenia (1533 km$^2$ catchment area), some 40 km before its discharge to the Adriatic Sea (Fig. 1). At this point, the mean annual discharge equals 94 m$^3$ s$^{-1}$ with monthly variations from 60 - 70 m$^3$ s$^{-1}$ in February and August to 110 - 120 m$^3$ s$^{-1}$ in May and June, and even 140 m$^3$ s$^{-1}$ in November. During the year, there are typically two flow extremes: a longer spring maxima from March to June (snowmelt) and a shorter but more intensive autumn maxima in October and November.

The first important tributary of the river Soča after Solkan is the river Vipava. The latter flows mainly through Slovenian territory and is hydrologically well elaborated in the cross-section at Miren on the Slovenian - Italian border (702 km$^2$ catchment area). The mean annual discharge at this point is 18 m$^3$ s$^{-1}$ with mean spring discharges of about 20 m$^3$ s$^{-1}$, which linearly decrease to only 7 m$^3$ s$^{-1}$ in August and increase again to the annual maxima of about 27 $m^3 s^{-1}$ in November and December.

The hydrology of the Italian part of watershed of the river Soča (known as the Isonzo in Italy) is less known as continuous measurements are not available downstream from Solkan. Moreover, a very complex system of surface and groundwater flows exists in a lower reach of the river Soča (MOSETTI, 1983). It comprises the eastern part of the alluvial Friuli plain with the river Torre in Italy, and the karstic area of the Kras at the Slovenian - Italian border, partly drained by the river Timavo. The river Torre is a tributary of the river Soča, while the river Timavo discharges directly into the Gulf of Trieste, in the area south of Monfalcone. The main problem is the character of the river Torre which, according to the saturation conditions of the plain, either loses or gains water along its flow. Up to present, the contribution of the Friuli plain to the river Soča hydrology was most relevantly estimated by analysis of various chemical properties of groundwater, and by analysis of the theoretical runoff based on the classical method of isohyets (MOSETTI, 1983). For our purpose, this estimation was generalised, so that the mean discharge at the river mouth is considered equal to 150 % of the sum of the mean discharges at Solkan and Miren. Although this is a rather rough simplification, such an approach is also consistent from the viewpoint of catchment surface areas (2235 $km^2$ in Slovenia and 1065 $km^2$ in Italy). The mean discharge of the river Soča at its mouth was thus set to 170 $m^3 s^{-1}$ which was somewhat more than the previously applied 150 $m^3 s^{-1}$ (ŠIRCA, 1996; ŠIRCA *et al.*, 1999) but agrees better with the values of other authors, e.g. 165 $m^3 s^{-1}$ (MOSETTI, 1983) and 172 $m^3 s^{-1}$ (BENINI, 1974).

Although with a lower reliability, the same relationship between discharges before confluence with the river Torre and at the river mouth was also used for high water events, e.g. as occurred in November 1997 (see below). These values were only roughly comparable to some estimations by MOSETTI (1983) who expected the peak discharges at the river mouth to reach as much as 3000 - 4000 $m^3 s^{-1}$. In November 1997, the peak discharge at the mouth was about 2500 $m^3 s^{-1}$.

## River sediment loads

The river Soča has a large transport capacity, which is due to its steep river bed slopes in the upper and middle reaches. These slopes increase from 0.138 % in a lower reach, to 0.198 % in the middle reach and even to 0.446 % along the river Idrijca. However, the transport capacity is affected by the reservoirs of 3 hydroelectric plants in Slovenia: the Doblar reservoir at the confluence with the river Idrijca (HPP Doblar), the small reservoir behind the Ajba dam providing water for the HPP Plave and the Solkan daily reservoir (HPP Solkan). It was assumed that these obstructions, especially the Doblar reservoir, prevent transport of all bedload originating from the area of Idrija and thus only mercury-rich suspended sediments can reach the Gulf of Trieste.

Our knowledge of the suspended sediment load of the river Soča suffered the same deficiency as has been mentioned with discharges, i.e. only few data from the Italian part of the river. Measurements in Slovenia were not available before the Kobarid measuring station some kilometres upstream from the confluence with the river Idrijca. Strong variations of daily average concentrations from a minimum of 1 g $m^{-3}$ to a maximum of 515 g $m^{-3}$ were recorded during the period of continuous measurements between 1966 to 1973, with a single peak reaching as much as 800 g $m^{-3}$. A rough estimate of the average concentration was 40 g $m^{-3}$. In the river Idrijca, up to three times higher peak concentrations (max 2574 g $m^{-3}$) were recorded by occasional measurements at the Hotešček measuring station since 1993. This suggested that the Idrijca might be a more turbid river than the upper Soča, which could partly be a consequence of uncontrolled runoff from the Idrija mine tailings.

For the period 1933-1935, a mean turbidity value of 150 g $m^{-3}$ was reported for the Soča at the cross-section of Kanal, some km upstream from Solkan (MOSETTI, 1983). The value is rather high and very probably far different from recent values, affected by dams. In October 1997, a value of 3 g $m^{-3}$ was measured at very low discharge in the river Soča at the bridge on the Monfalcone-Grado road, immediately upstream of the river Soča mouth. Another measurement at very high water in November 1997 gave a mean concentration of 365 g $m^{-3}$ at the same site (HORVAT *et al.*, in press). The latter value was highly relevant for cases of extreme episodic events, but was not applicable to average conditions. For the Vipava and the Torre tributaries, only some unreliable estimates exist in the older literature. As for the Idrijca, they were often expressed in $m^3$ $year^{-1}$, which gave little information about actual suspended sediment concentrations.

However, except for the Kobarid measuring site, the data on suspended sediment were not sufficient to give a reliable estimate of annual loads to the Gulf. For the purpose of the mercury mass balance, an average river suspended sediment concentration of 28 g $m^{-3}$ was estimated on the basis of sediment deposition rates in the vicinity of the mouth and by balancing with other, more reliable terms of the mercury budget. Two comparable mean annual values from the literature were 130 g $m^{-3}$ for the river Adige in northern Italy (JURAČIĆ *et al.*, 1987) and 40 g $m^{-3}$ for the river Loire in France (RELEXANS *et al.*, 1988). Although these two values indicate a possible underestimation in our case, the value of 28 g $m^{-3}$ was finally kept as a reliable minimum for the purpose of the mercury mass balance. In any case, extensive further research is necessary (and planned) on this topic.

## Mercury concentrations

Mercury concentrations at the mouth of the river Soča as an input to the marine environment of the Gulf of Trieste were measured during four well documented (Table 1) and two further measuring campaigns (SEADATA S.R.2, 1992) since 1990. It was obvious from these measurements that the majority of mercury entered the Gulf in particulate form as concentrations in solution were low even during the very intensive flood in 1997 (see also next section). The average value of river Soča dissolved mercury applied in the final mass balance, 1.6 ± 1.0 μg $m^{-3}$, was calculated from the measurements of all six campaigns. The share of MMHg in this amount was determined from the two most recent campaigns (1997, 1998) and averaged to 1.65 %.

On the other hand, concentrations of particulate mercury varied in a range from less than 10 to almost 80 mg $g^{-1}$ d.w. with higher values appearing during high water events. They were

*Table 1. Mercury in water and suspended matter of the river Soča (HORVAT* et al., *in press)*

| site | sampling location | date | estimated discharge | Hg tot | Hg diss | MMHg tot | MMHg diss | susp. matter |
|---|---|---|---|---|---|---|---|---|
| | | | $m^3s^{-1}$ | ng $l^{-1}$ | ng $l^{-1}$ | ng $l^{-1}$ | ng $l^{-1}$ | $gm^{-3}$ |
| mouth | surface | 19.12.90 | 101 | 8.7±1.0 | - | - | - | - |
| | bottom | 19.12.90 | 101 | 6.4±0.9 | - | - | - | - |
| mouth | surface | 17.06.92 | 107 | 14.5 | - | - | - | - |
| | bottom | 17.06.92 | 107 | 10.6 | - | - | - | - |
| bridge | left | 08.11.97 | 1700 | 15825 | 3.49 | 0.314 | 0.048 | 260 |
| (mouth) | right | 08.11.97 | 1700 | 17999 | 2.47 | 0.285 | 0.039 | 470 |
| border | n.a. | 23.11.98 | 50 | 1.5 | 1.26 | 0.048 | 0.023 | 12.4 |
| bridge | right | 23.11.98 | 85 | 1.8 | 1.27 | 0.052 | 0.022 | 5.1 |
| (mouth) | middle | 23.11.98 | 85 | 3.66 | 1.50 | 0.010 | 0.005 | 36.2 |

thus supposed to be well correlated with the river discharge but this was not yet confirmed. As some measurements in 1991 (HORVAT *et al.*, 1996) showed an average concentration of 7.5 mg $g^{-1}$ in the surface layer of the bottom sediment and some more recent surveys confirmed this average with values around 10 mg $g^{-1}$, the value adopted for the mass balance was 10 mg $g^{-1}$ d.w. The share of MMHg was as low as < 0.1 % during flood and around 3 % during average flow conditions. In the mass balance, the value of 0.1 % was adopted for MMHg.

## The 1997 autumn flood wave

In the beginning of November 1997, a flood wave of great intensity was recorded in the river Soča watershed. The peak value of discharge at the Solkan measuring station was 1642 $m^3s^{-1}$, which corresponded to the discharge of a 5-year return period ($Q_{05}$). Suspended matter concentrations were measured at the Hotešček measuring station on the river Idrijca and at the bridge on the Monfalcone-Grado road (Table 2). At the latter location, samples for mercury concentration determination in water and suspended matter were also taken (Table 3).

The basis for the calculation of mercury input to the Gulf during this flood event was the continuous time-series of discharges at the river Soča mouth. As the suspended matter concentrations had not been monitored continuously, they were estimated from discrete measured values (Table 2). To this end, an average flow velocity of 2.8 m $s^{-1}$ was applied for the Hotešček - Solkan reach and 1.75 m $s^{-1}$ for the Solkan - mouth reach. Both values applied for the $Q_{05}$ conditions and gave a water travel time of 2.7 hours between Hotešček and Solkan and 6.5 hours between Solkan and the mouth.

Calculations of mercury loads during the flood wave (Table 4) confirmed the previously assumed importance of episodic events for the mercury mass balance in the Gulf. In more than 600 million $m^3$ of water discharged into the Gulf during 8 flood days, the total amount of mercury reached 4700 kg. This was more than triple the value of the estimated average annual gain from the river Soča (see mass balance below). On the other hand, the total amount of MMHg discharged by the flood was about 100 g. This resulted from the average concentration of 44 ± 9 pg $l^{-1}$ of dissolved MMHg and 0.77 ± 0.27 ng $g^{-1}$ of particulate bound MMHg.

*Table 2. Suspended matter concentrations in the river Soča watershed during the 1997 autumn flood wave ( * denote estimated values)*

| stream | location | date | time | discharge | susp. matter |
|---|---|---|---|---|---|
| | | | | $m^3s^{-1}$ | $gm^{-3}$ |
| *Idrijca* | *Hotešček* | *1997-11-07* | *07:00* | *174* | *88* |
| | | | *09:45* | *190* | *70* |
| | | | *13:00* | *311* | *176* |
| | | | *18:00* | *222* | *170* |
| *Idrijca* | *Hotešček* | *1997-11-08* | *07:00* | *337* | *207* |
| | | | *15:00* | *225* | *565* |
| *Idrijca* | *Hotešček* | *1997-11-09* | *07:00* | *109* | *37* |
| | | | *18:00* | *92.5* | *57* |
| *Soča – Isonzo* | *near mouth - left* | *1997-11-08* | *12:00* | *1700** | *260* |
| | *near mouth - right* | | *12:00* | *1700** | *470* |

*Table 3. Mercury concentrations in the river Soča water and particulate matter during the 1997 autumn flood wave. Sampling site was the bridge on the Monfalcone - Grado road*

| location | dissolved | | particulate | | susp. matter |
|---|---|---|---|---|---|
| | $Hg_{TOT}$ | MMHg | $Hg_{TOT}$ | MMHg | |
| | $ngl^{-1}$ | $ngl^{-1}$ | $\mu gg^{-1}$ | $ngg^{-1}$ | $gm^{-3}$ |
| left bank | 3.42 | 0.041 | 78.12 | 1.177 | 260 |
| | 3.56 | 0.055 | 43.58 | 0.865 | 260 |
| right bank | 2.48 | 0.031 | 30.49 | 0.504 | 470 |
| | 2.46 | 0.047 | 46.09 | 0.543 | 470 |

*Table 4. Mercury loads to the Gulf of Trieste during the river Soča flood wave in November 1997*

| | upstream of the Torre | downstream from the confluence with the river Torre (the river Soča mouth) | | | | |
|---|---|---|---|---|---|---|
| date | daily water volume [$10^6$ $m^3$] | daily water volume [$10^6$ $m^3$] | cumulative water volume [$10^6$ $m^3$] | estimated mean susp. matt. conc. [$gm^{-3}$] | daily amount of susp. matt. [t] | **daily amount of mercury [kg]** |
| 1997-11-06 | 20 | 30 | 30 | 50 | 1500 | **75** |
| 1997-11-07 | 108 | 162 | 192 | 120 | 19440 | **972** |
| 1997-11-08 | 100 | 150 | 342 | 400 | 60000 | **3000** |
| 1997-11-09 | 58 | 87 | 429 | 50 | 4350 | **218** |
| 1997-11-10 | 55 | 82 | 511 | 50 | 4100 | **205** |
| 1997-11-11 | 34 | 51 | 562 | 50 | 2550 | **128** |
| 1997-11-12 | 29 | 44 | 606 | 50 | 2200 | **110** |

## BENTHIC MERCURY FLUXES

Mercury fluxes at the bottom of the Gulf were determined by a benthic chamber experiment at location AA1 (Fig. 2) during 1995 and 1996 (COVELLI *et al.*, 1999b). According to this study, the downward flux of mercury was 2.96 mg $m^{-2}year^{-1}$. The net sedimentation (burial) was only 2.18 mg $m^{-2}$ $year^{-1}$, while the remaining 0.78 mg $m^{-2}year^{-1}$ was resuspended. MMHg represented only 1.8 % of total Hg (0.04 mg $m^{-2}$ $year^{-1}$) in the buried segment but 24 % (0.19 mg $m^{-2}$ $year^{-1}$) in resuspension (COVELLI *et al.*, 1999b). The total downward flux was also comparable to results of a sediment trap experiment in the southern part of the Gulf where an intensity of 3.10 mg $m^{-2}$ $year^{-1}$ was obtained at a depth of 20 m, and an intensity of 1.29 mg $m^{-2}$

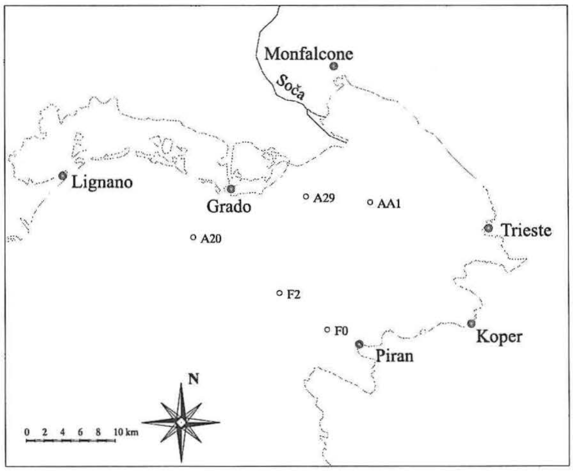


*Fig. 2. Sampling locations for benthic fluxes (COVELLI et al., 1999b) and open boundary fluxes*

year$^{-1}$ at a depth of 10 m (PLANINC & FAGANELI, 1993). Furthermore, the net downward flux after COVELLI *et al.* was well comparable to the average mercury sedimentation rate of 2.77 mg m$^{-2}$ year$^{-1}$, obtained by the 2D STATRIM mercury cycling model (ŠIRCA, 1996; ŠIRCA *et al.*, 1999).

If we apply the rates of COVELLI *et al.* (1999b) to the whole area of the Gulf (approx. 600 km$^2$), 1776 kg of Hg would be deposited annually on the sea bottom. The net deposition (burial) would amount to 1308 kg, which is in an acceptable agreement with the previously estimated value of 1666 kg (ŠIRCA, 1996; ŠIRCA *et al.*, 1999). Most importantly, the downward MMHg flux of 24 kg year$^{-1}$ and the upward flux of 114 kg year$^{-1}$ mean that the sea bottom of the Gulf is the most important source of MMHg for the Gulf, with the magnitude of 90 kg of MMHg per year.

## OPEN BOUNDARY FLUXES

Determination of mercury fluxes from and to the open sea of the Northern Adriatic was based on assumption of a counter-clockwise residual flow in the Gulf (ŠIRCA *et al.*, 1999; ŠIRCA and RAJAR, 1997a, 1997b) and on measurements of mercury for open boundary inflow at points F0 and F2 (Fig. 2), and for open boundary outflow at points A29 and A20, with Hg concentrations as reported by HORVAT *et al.* (in press). The magnitude of the inflowing stream at the southern coast of the Gulf was 1500 m$^3$ s$^{-1}$, while it increased to an outflow of 1670 m$^3$ s$^{-1}$ at the northern coast, due to the addition of the average discharge of the river Soča.

The average inflowing concentration of total marine water mercury was 1.6 ± 0.7 ng l$^{-1}$ with 3.6 % of the amount being MMHg in near-bottom samples. As MMHg was under the detection limit in inflowing surface samples, its share was assumed to be negligible in the total mercury mass balance. The average outflowing mercury concentration was 5.4 ± 2.4 ng l$^{-1}$ with 0.5 % of this amount being MMHg. There were again considerable differences between bottom (0.8 % of MMHg) and surface (0.2 % of MMHg) samples but the application of the mean value was based on assumption of a strong vertical mixing over the shelf along the northern coast.

## ATMOSPHERIC MERCURY

Wet deposition of mercury from the atmosphere was determined through a mean annual rainfall of 1050 l m$^{-2}$ and mercury content of 6 ng l$^{-1}$. The average daily load of about 17 ng m$^2$day$^{-1}$ was comparable to, although lower than, some measured values from Scandinavia (e.g. IVERFELDT *et al.*, 1995; MUKHERJEE *et al.*, 1995). The total wet deposited load of atmospheric mercury to the Gulf was 3.8 ± 1.9 kg year$^{-1}$. The proportion of MMHg in this amount was not yet determined, nor were the fluxes of elemental mercury at the water - air interface (evasion). Further work is in progress.

## DISCUSSION

**Gains.** Because the measured data were scanty and of low reliability, the average river Soča sediment concentration was calculated from the estimated annual sediment load to the Gulf and set at 28 g m$^{-3}$. The average particulate mercury concentration was 10 mg g$^{-1}$ d.w. Mercury fluxes from the sea bottom were determined by a benthic chamber experiment at a location in the middle of the Gulf during 1995 and 1996 (COVELLI *et al.*, 1999b), where also intensive resuspension was confirmed. At the southern part of the open boundary, a residual stream of 1500 m$^3$ s$^{-1}$ brings in relatively clean water with 1.6 ± 0.7 ng l$^{-1}$ total Hg concentration. Estimation of the Hg input from the atmosphere was 6 ± 3 ng l$^{-1}$ in rainwater. The total amount of incoming mercury was 1591 kg per year.

**Losses.** In the benthic chamber, the total downward flux of mercury, 2.96 mg m$^{-2}$ year$^{-1}$, was comparable to earlier results of a sediment trap experiment in the southern part of the Gulf (PLANINC & FAGANELI, 1993), and to the computed average mercury burial rate (ŠIRCA *et al.*, 1999). However, it remains unclear whether the benthic chamber location is representative of the whole Gulf bottom. A 1670 $m^3$ $s^{-1}$ residual stream through the northern part of the open boundary carried out waters with Hg concentrations of 5.4 ± 2.4 ng $l^{-1}$. Evasion to the atmosphere was not yet measured nor calculated but it is a possibly significant term of the balance. In view of some literature data (EBINGHAUS *et al.*, 1999; LAMBORG *et al.*, 1999), its magnitude is not expected to be less than 100 kg $year^{-1}$. The total amount of outgoing mercury was 1592 kg per year.

**Mass balance.** On the basis of presently available measurements it was possible to balance the estimated sources and sinks as presented in Fig. 3, where most of the items of the budget were expressed with expected (±) tolerances. The values of MMHg in brackets (Fig. 3) represent the share of this form in respective amounts of total Hg. It was assumed in the mass balance that 26 % (468 kg) of settled mercury re-entered the water column due to resuspension and due to diffusion at the sea bed - water interface, but did not leave the Gulf. It can be speculated that most of the net MMHg flux from the sea bottom (114 - 24 = 90 kg $year^{-1}$) enters various food chains and is thus either continuously accumulated in biota or removed

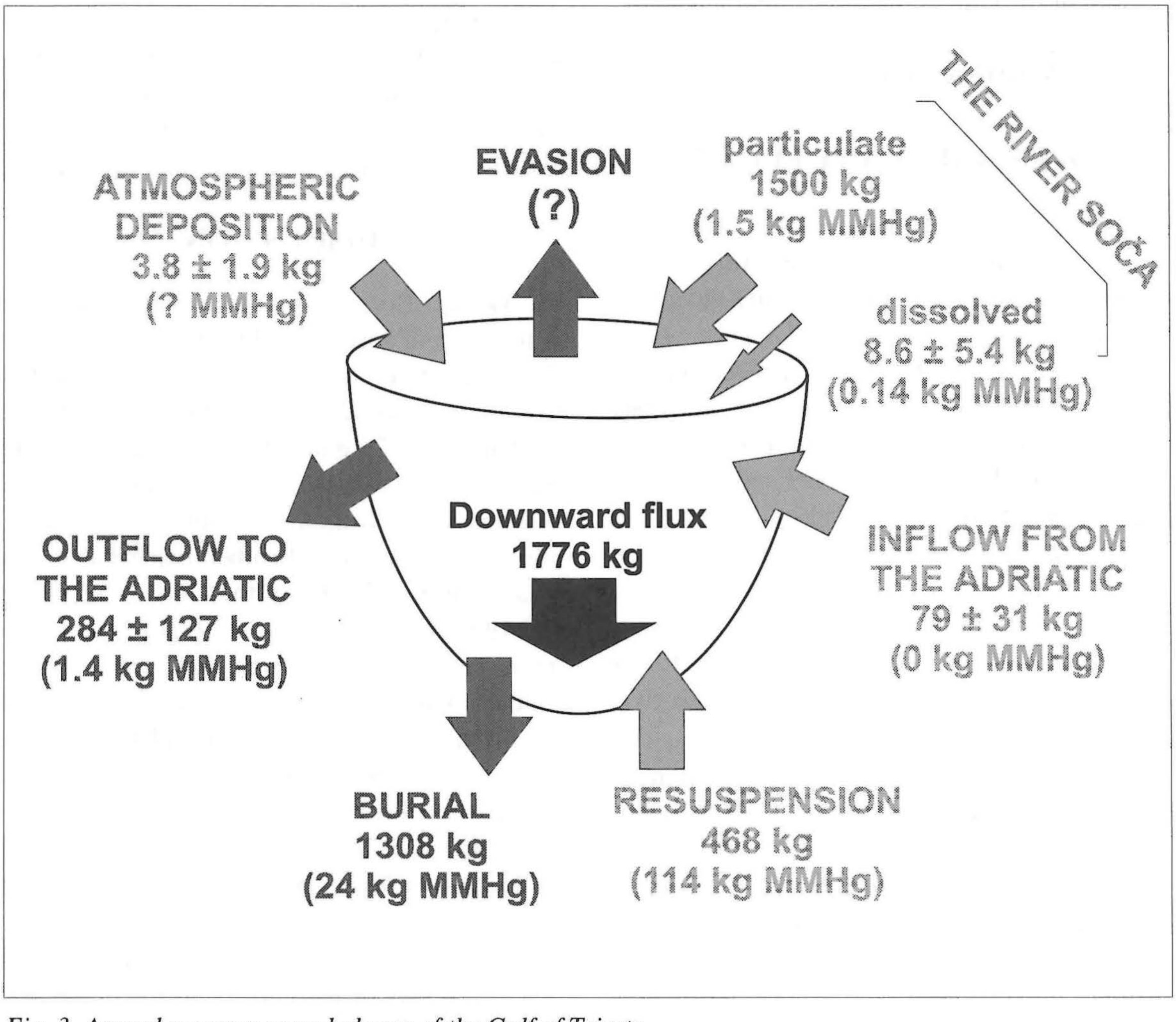


*Fig. 3. Annual mercury mass balance of the Gulf of Trieste*

from the marine environment by fishing and fish migration.

However, the three most questionable items remained: 1) the annual suspended sediment yield from the river Soča catchment, 2) the Hg burial rate determined only at one location in the Gulf and 3) possibly underestimated outflows through the open boundary, which might occur during exceptional events of high winds with not yet recorded mercury concentrations.

## CONCLUSIONS

The present knowledge of mercury fluxes in the Gulf of Trieste only enables a provisional mass balance. Since the establishment of the first approximate mass balance in 1996, the (then assumed) strong burial rate was confirmed by an *in situ* experiment and the sea bottom sediments were identified as the main methylmercury source for the Gulf. Extensive future research is planned for evaluation of sediment transport from the river Soča catchment to the sea. Also planned are further benthic chamber experiments at other locations, and measurements for determination of the fluxes at the air-water interface. Possible exceptional sediment-displacing events are being elaborated in the framework of development of a 3D sediment transport model of the Gulf (ŽAGAR *et al.*, 1999).

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# Procjena bilance mase žive u Tršćanskome zaljevu

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**SAŽETAK**

Predstavljena je godišnja bilanca mase žive u Tršćanskome zaljevu, koji kao izvore uključuje dotok rijekom Sočom, iz otvorenoga mora Sjevernog Jadrana, iz atmosfere u obliku oborina i morskoga dna putem resuspenzije. Živa opada u području otjecanjem u otvoreno more i taloži se na dno. Dotok rijekom Sočom (oko 1500 kg Hg godišnje) i taloženje (oko 1300 kg Hg godišnje) su dva glavna čimbenika ove bilance. Unatoč snažnoj resuspenziji, ukupno 82 % žive ukloni se godišnje iz vodenog stupca taloženjem. S druge strane, morski su sedimenti glavni izvor monometilne žive (MMHg) u zaljevu, koja na kontaktnoj plohi sediment-voda ulazi u vodeni stupac intenzitetom od 90 kg MMHg godišnje.