

Acid-labile sulfides in shallow marine bottom sediments: A review of the impact on ecosystems in the Azov Sea, the NE Black Sea shelf and NW Adriatic lagoons

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ABSTRACT

Acid-labile sulfides (LS) increase in bottom sediments at sites in the Azov Sea, at the NE Black Sea shelf and in the coastal lagoons of NW Adriatic Sea experiencing direct impacts of anthropogenic pollution. Fresh anthropogenic organic matter stimulates the bacterial sulfate reduction and here the rate of the LS production overcomes their loss during the oxidation and pyritization. This results in the expansion of reduced sediment layer up to the bottom surface. The LS concentration in the reduced sediments varies between 300 and 2000 mg S l⁻¹ of wet silt depending on the size of pollution loading and on the rate of sedimentation. In the oxidized sediments away from the direct pollution impact, the LS concentration did not exceed 100–150 mg S l⁻¹. Being a strong cytochrome toxin, the LS adversely affect the coastal ecosystems. The concentrations over 600 mg S l⁻¹ result in quasi total benthic mortality whereas >300–400 mg S l⁻¹ depletes the benthic faunal abundance and taxonomic diversity. Accumulation of the LS in sediments also induces nocturnal hypoxia and stimulates domination of toxic cyanobacteria in the pelagic phytocenoses.

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1. Introduction

Labile (acid soluble) sulfides (LS) are formed in anoxic biotopes as the result of anaerobic processes of biogenic sulfidogenesis (Sorokin, 1960; Jorgensen, 1977b, 1981; Bussman and Reichart, 1991) especially in the layer of reduced bottom sediments. These sediments have a darkish or black color because of accumulation of the black iron monosulfide (hydrotroilite) although under the iron deficiency, the layer of reduced sediments might be greenish or may retain a gray color. In this case, this layer might be recognized by the elevated concentrations of LS (>200 mg S l⁻¹ of wet sediment). In the basins with oxic water column, the depth position of reduced layer within the sediment column depends on the balance between the rates of sulfide production and its loss (Volkov, 1961). If the production exceeds the loss, the layer of reduced sediment may reach the bottom surface. This happens in the coastal and estuarine zones subject to anthropogenic pollution due to the increased loading of fresh organic matter, which is the factor critically controlling the rate of sulfidogenesis (Sorokin, 1960; Jorgensen, 1977b; Chebotarev et al., 1983; Stankhowitch, 1984; Sorokin et al., 1999a). Therefore, in areas experiencing most intense pollution impact, the layer of reduced sediments usually

appears at the bottom surface (Jorgensen, 1977b; Bussman and Reichart, 1991; Sorokin et al., 2002; Han et al., 2008).

The concentration of LS in the reduced sediments at the marine shelf with redox potential below 100 mV usually varies from 200 to 1000 mg S l⁻¹ of wet mud, or from 0.04 to 0.2% of its dry weight. Oxidized muddy shelf sediments located in the upper sediment layer may also contain up to 150 mg S l⁻¹ of LS. Free sulfide migrates here from the underlying reduced sediments and is also forming there in the anaerobic microniches (Jorgensen, 1977a; Sorokin et al., 1999a, 2002).

Sulfide is known as a strong cytochrome toxin (Giere, 1992; Gamenick and Theede, 1996; Gamenick et al., 1996; Como and Magani, 2002). Sulfide accumulation in upper sediment layer in coastal and estuarine areas receiving direct organic pollution exerts an impact upon not only local benthic but also the pelagic biota due to the release of free sulfide into the water column (Giordani, 1991). Being oxidized with the dissolved oxygen, it enhances the deoxygenation of the water column thus inducing nearbottom hypoxia even by a slight water column stratification (Fedorov, 1955; Grishaber et al., 1992; Yakushev et al., 2003). The hypoxia enhances the inhibiting impact of sulfide upon the local biota leading to a severe transformation of the whole coastal ecosystem in such anthropogenically stressed areas, and a loss of their catchable resources (Stankhowitch, 1984; Rosenberg and Loo, 1988; Giere, 1992; Sorokin et al., 2002, 2006; Papadas et al., 2009). Here we hypothesize that there is an increasing LS concentration in the

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shelf zone of N–E Black Sea, in the shallow Azov Sea and in some lowland lagoons of the N–W Adriatic experiencing intense pollution stress leading to the increased impact of sulfide accumulation upon the coastal ecosystems.

2. Material and methods

The LS concentration was measured in over 200 samples from the upper (0–4 cm) layer of soft bottom sediments in 2002–2004 in the Azov Sea and in the NE shelf of the Black Sea, and in 1995–2001 in the lagoons of the NW Adriatic Sea coast. The sediment samples were collected into 200 ml glass containers from monoliths taken with the Eckmann-Bergays box grab, fixed with Zn-acetate alkaline solution and were closed without an air bubble. The concentration of LS was analyzed in the sediment samples treated with 10% sulfuric acid in the apparatus shown in Fig. 1. The distillation of H_2S from the acidified sediment samples was accomplished as follows. A sediment subsample (15–20 ml) taken with the calibrated tube was carefully mixed with 20 ml of 5% sodium bicarbonate solution + 2–3 ml of isobutanol added to preventing foaming. This slurry was transferred into the reaction tube. The absorber tube was filled with the mixed solution of $CdSO_4$ (2%) + Zn-acetate (0.5%) and +0.1 ml KOH solution (0.05%). Then the flux of nitrogen gas was introduced into the apparatus and 5% solution of sulfuric acid is added by drops to acidify the slurry. After 10 min 20 ml of 10% sulfuric acid was added and the solution distilled for 20–30 min. The sulfide concentration was assessed then iodometrically in the absorbing solution.

3. Results

3.1. North-eastern shelf of the Black Sea

The LS concentration was measured at 80 stations situated between the Kerch strait to the Khosta resort city including the Zemess bay of the Novorossiisk port and the bays of Gelendzhik and the Tuapse port cities (Tables 1 and 2). The highest LS concentration in the upper sediment layer was observed in the shelf sites situated near to large recreation settlements and also in the bays harboring port cities. The LS concentration in such areas ranged from 300 to 980 $mg\ S\ l^{-1}$ of wet silt and from 100 to 170 $mg\ S\ l^{-1}$ in the sandy sediments. This means that in the shelf zones subject to direct pollution impact from the resort and also from the port cities the layer of reduced sediment with high LS concentration increases in the bottom surface. The LS concentration reaches its maximum values of 700–900 $mg\ S\ l^{-1}$ in the sediments of the port bays subject to intense pollution arriving with the municipal, port and agriculture waste waters. Those levels of LS in the bottom sediments are comparable to those in the coastal marine environments experiencing the most intense external pollution (Sorokin and Bilio, 1981; Bussman and Reichart, 1991; Gamenick et al., 1996; Sorokin et al., 1999a, 2002, 2006; Heijs et al., 2000).

High LS concentrations of between 500 and 950 $mg\ S\ l^{-1}$ were recorded in the soft shelf sediments collected opposite Idocopas in the zone accepting wastewater from the large recreation settlements of Arkhipo-Osipovka and Divnomorsk (Table 1). Reduced sediments with sulfide concentrations of 300–500 $mg\ S\ l^{-1}$ were also observed in the shelf zones of 20–50 m deep situated opposite the large recreation settlements such as Khosta, Lazarevskoe, Pshada, Ozereevka. The shelf zones situated outside the direct pollution impact were occupied by oxidized sediments with the LS concentration in the upper layer less than 200 $mg\ S\ l^{-1}$. This level is usual in oxidized sediments containing a rich bottom flora and fauna (Jorgensen, 1977a). The sandy sediments with a moderate LS concentration of 150–250 $mg\ S\ l^{-1}$ were recorded in the

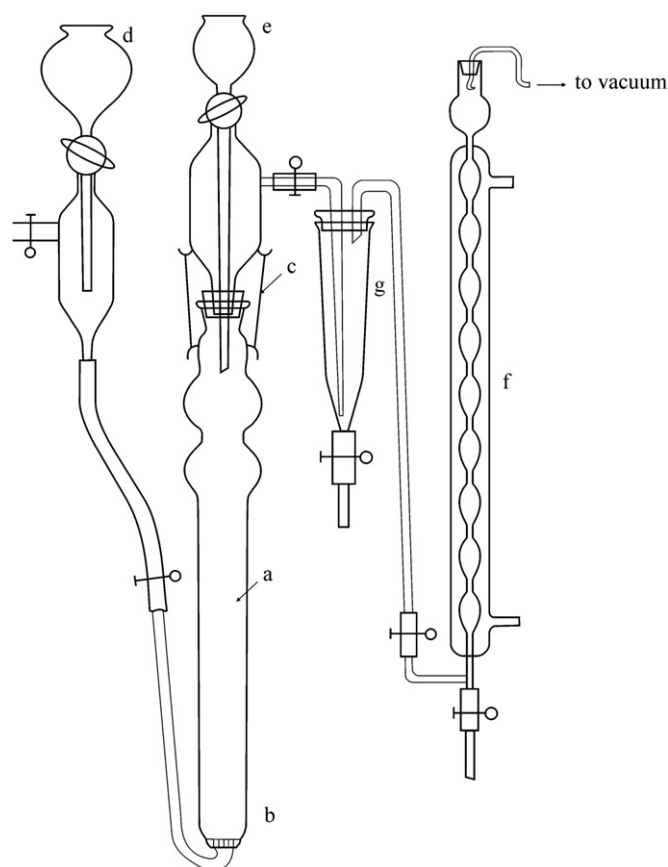


Fig. 1. Scheme of apparatus for distillation of H_2S from the sediments: a – reaction tube; b – porous glass plate; c – rubber bands to fix in the reaction tube; d – funnel for adding the acid; e – funnel for adding the 2-butanol antifoam mixture; f – absorber; g – emergency foam trap.

Gelendzhik and in the Zemess bays in zones of an intense sedimentation, where the LS was diluted by the mineral fraction.

The rate of sulfate reduction was assessed in the samples collected in the Gelendzhik and the Zemess bays, and in adjacent shelf zone (Table 3). The maximal rates were recorded in the sites experiencing direct pollution impact from the port and from the cities situated along their shores. The rate of sulfate reduction varied there from 10 to 44 $mg\ S\ (l\ d)^{-1}$ of wet silt or 300–1300 $mg\ S\ d^{-1}\ m^{-2}$ of the bottom surface. These rates are comparable with those reported for the sediments from eutrophic and hypereutrophic shallow marine basins experiencing high level of anthropogenic pollution (Tolokonnikova, 1977; Sorokin and Bilio, 1981; Sorokin et al., 1996a).

There was a pronounced negative impact of sulfide accumulation in the upper sediment layer upon abundance and diversity of zoobenthos (Table 1) (Theede, 1973; Giere, 1992). The LS accumulation over 500 $mg\ S\ l^{-1}$ of wet silt results in an overall benthic mortality (Stankhowitch, 1984). In our case, the biomass of zoobenthos in such sediments found in zones of direct pollution impact decreased to $<2\ g\ m^{-2}$, being represented by some predatory polychaetes such as *Nereis*, *Capitella*, *Polydora* and by the bivalve *Tritia*, which are tolerant to sulfide (Vismann, 1991; Grishaber et al., 1992; Gamenick and Theede, 1996). In reduced sediments with the LS concentration 300–480 $mg\ S\ l^{-1}$, the zoobenthos showed a depleted biomass ($<20\ g\ m^{-2}$) and diversity evidenced by the domination of taxa tolerant to sulfides. The benthic communities showing their quasi-normal state were found only in zones occupied by the oxidized sediments outside of direct

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