

# Soil Iodine Determination in Deccan Syncline, India: Implications for Near Surface Geochemical Hydrocarbon Prospecting

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The association of iodine with organic matter in sedimentary basins is well documented. High iodine concentration in soils overlying oil and gas fields and areas with hydrocarbon microseepage has been observed and used as a geochemical exploratory tool for hydrocarbons in a few studies. In this study, we measure iodine concentration in soil samples collected from parts of Deccan Syncline in the west central India to investigate its potential application as a geochemical indicator for hydrocarbons. The Deccan Syncline consists of rifted depositional sites with Gondwana–Mesozoic sediments up to 3.5 km concealed under the Deccan Traps and is considered prospective for hydrocarbons. The concentration of iodine in soil samples is determined using ICP-MS and the values range between 1.1 and 19.3 ppm. High iodine values are characteristic of the northern part of the sampled region. The total organic carbon (TOC) content of the soil samples range between 0.1 and 1.3%. The TOC correlates poorly with the soil iodine ( $r^2 < 1$ ), indicating a lack of association of iodine with the surficial organic matter and the possibility of interaction between the seeping hydrocarbons and soil iodine. Further, the distribution pattern of iodine compares well with two surface geochemical indicators: the adsorbed light gaseous hydrocarbons (methane through butane) and the propane-oxidizing bacterial populations in the soil. The integration of geochemical observations show the occurrence of elevated values in the northern part of the study area, which is also coincident with the presence of exposed dyke swarms that probably serve as conduits for hydrocarbon microseepage. The corroboration of iodine with existing geological, geophysical, and geochemical data suggests its efficacy as one of the potential tool in surface geochemical exploration of hydrocarbons. Our study supports Deccan Syncline to be promising in terms of its hydrocarbon prospects.

**KEY WORDS:** Light gaseous hydrocarbons, propane-oxidizing bacteria (POB), TOC, Mesozoic, dykes.

## INTRODUCTION

The distribution of iodine in earth crust varies, with sedimentary rocks having higher values com-

pared with the igneous and metamorphic rocks (Muramastu and Wedepohl, 1998). The iodine content in sedimentary rocks show a broad range from <0.1 to 150 ppm. The average value in shale, sandstone, and carbonate rocks is 2.3, 0.8, and 2.7 ppm, respectively (Fuge, 1996). A large number of studies show the organic matter as major concentrator of iodine in sedimentary basins, and values as high as 44 ppm are reported (Vinogradov, 1959; Collins and Egleeson, 1967; Collins, 1969; Cosgrove, 1970;

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Whitehead, 1973, 1978; Ullman and Aller, 1980; Sheppard and others, 1995; Fuge, 1996; Moran, 1996; Fehn, Snyder, and Muramatsu, 2007). Iodine concentrations correlate well with changes in organic matter of marine sediment, both at a given location and from location to location (Price and Calvert, 1973; Pedersen and Price, 1980). Kartsev and others (1959) reported low concentration of iodine in fresh surface and ground waters ( $10^{-5}$  to  $10^{-3}$  ppm); sea water ( $5 \times 10^{-2}$  ppm); and underground saline waters ( $10^{-1}$  to 1 ppm) in non oil-bearing regions, whereas it was found to be more concentrated in fresh waters of oil-bearing sediments by a factor of  $10^4$  to  $10^7$ . Enrichment of iodine in oil field brines from sedimentary basins is well established, and the concentration ranges between 10 and 100 ppm on an average (Collins and Egleeson, 1967; Moran, 1996). Iodine as high as 1560 ppm in the brines of Anadarko basin has been reported (Collins, 1969; Moran, 1996; Moran, Fehn, and Ray, 1998). These concentrations are significantly higher when compared with any other geologic material and fluid (rock-forming minerals and evaporites =  $\ll 10$  ppb; sea water =  $\sim 0.05$  ppm of iodine). The seaweeds typically concentrate 100–5000 ppm of iodine (Vinogradov, 1953; Collins, 1969; Wong, Brewer, and Spencer, 1976). The iodine in brines of sedimentary basins with large accumulations of hydrocarbons is said to be derived from the organic matter that accumulated in the subsiding basin and the subsequent concentration by dewatering and compaction of sediments (Collins, 1969; Collins, Bennett, and Manuel, 1971; Moran, 1996). The iodine concentration in the connate waters has been used as source indicator for hydrocarbons for several decades (Collins and Egleeson, 1967; Collins, 1969; Moran, 1996). The strong association of iodine with organic material makes the use of iodine isotopic system useful in tracing and dating organic materials and their derivatives, including the hydrocarbons (Fehn, Snyder, and Muramatsu, 2007; Tomaru and others, 2007).

The unique relation of iodine with the organic matter has been utilized by several workers in surface geochemical hydrocarbon prospecting (Kartsev and others, 1959; Allexan and others, 1986; Tedesco and others, 1987; Tedesco, 1995; Leaver and Thomasson, 2002). The surface geochemical survey takes into account the microseepage of light gaseous hydrocarbons (methane through butane) from the subsurface reservoirs to the shallow surface soils, resulting in various surface manifestations indicative

of the subsurface accumulations (Jones and Drozd, 1983; Xuejing and Binzhong, 1989; Klusman, 1993; Schumacher and Abrams, 1996). The vertical/near vertical migration of the hydrocarbon induces chemically identifiable changes in the surface soils and one such altered parameter is the soil iodine concentration. The iodine concentrations are observed to exceed several times the background value in soils overlying the microseepage areas (Kartsev and others, 1959; Allexan and others, 1986; Tedesco and others, 1987; Tedesco, 1995). The use of iodine as an exploratory tool for hydrocarbons has salient advantages of simplicity and cost effectiveness apart from its dependable and consistent results. Also, the short-term fluctuations due to diurnal variations have minimum accountability in the results, thus constraining the exploration risk and cost to a considerable extent (Leaver and Thomasson, 2002).

However, the lack of the sufficient published literature in diverse geological regions necessitates the need for a detailed research work laying emphasis on the application of iodine as one of the natural integrators of hydrocarbons. The article presents the geochemistry of iodine in terms of its concentration distribution in surface soils, its relation with the surface organic matter and its implication for oil and gas prospects from a reconnaissance survey carried out in parts of Deccan Syncline (DS), west central India. The region has received wide attention for its hydrocarbon prospects because of the thick Gondwana–Mesozoic sedimentation of 2–3.5 km buried under the lava flow of Deccan Traps and the article delves further to identify the possible association of iodine with hydrocarbon seepage. The result of soil iodine studies have been substantiated by adsorbed light gaseous hydrocarbon data ( $C_1$ – $C_4$ ), propane oxidizing bacterial populations, total organic carbon contents and the geological and geophysical studies for the hydrocarbon occurrences in the area.

#### **EARLIER WORK ON USE OF IODINE AS AN INDICATOR OF HYDROCARBON**

Iodine has been suggested as hydro-geochemical indicator for oil and gas by Russians nearly five decades ago. Kartsev and others (1959) reported high iodine concentration in the fresh waters of oil-bearing sediments by a factor of  $10^4$  to  $10^7$  and suggested that major part of iodine in waters of oil-bearing sediments came from oil, the probable cause

being the seaweeds which concentrated the element. Kovda and Salvin (1951) showed that the normal iodine content in soils is of order of  $10^{-4}\%$  whereas the iodine content of soils over the oil formations reaches  $10^{-3}\%$  or  $10^{-2}\%$ . In recent past, case histories were published using iodine as an exploratory tool from Thrace basin of Turkey and several petroleum producing basins of USA viz. Denver basin in Colorado and Nebraska; Keota Dome in Iowa basin; Illinois basin in Illinois; Forest City basin in Kansas; Midland basin in Texas; Powder River Basin in Wyoming and Williston basin in North Dakota (Allexan and others, 1986; Tedesco and others, 1987; Leaver and Thomasson, 2002; Goudge, 2007, 2009). The workers have used iodine surveys as reconnaissance as well as detailed developmental tool depending on the objective of exploratory survey (Allexan and others, 1986; Tedesco and others, 1987).

The mechanism and the chemical form in which iodine occurs in soils overlying hydrocarbon reservoirs or microseepage areas are poorly understood. Kartsev and others (1959) categorized iodine as one of the direct hydro-geochemical indicator because it entered the water from the water soluble component of oil due to the dissolution in salt or ionic form. Vertical migration of the subsurface brines containing high concentration of iodine toward shallow surface in the sedimentary basin is considered as one probable cause for high iodine values in surface soils of petroleum accumulations (Collins and Egleeson, 1967; Fabryka-Martin and others, 1985; Fabryka-Martin, Davis, and Elmore, 1987; Moran, Fehn, and Hanor, 1995). Evidences of upward flow of fluids in sedimentary basins toward shallow surface has been demonstrated by Land (1991), which include natural seepage of oil and gas, and presence of allochthonous brines in areas where deep seated growth faults penetrate the sedimentary sections. Dense brine has been reported to migrate through the sediments over a distance of >500 km (Martin and others, 1993; Moran, Fehn, and Hanor, 1995; Moran, Fehn, and Ray, 1998). The source of iodine in brines of sedimentary basins with large accumulations of hydrocarbons is suggested to be the organic matter which, after burial and maturation in suitable conditions, becomes precursor to crude oil. (Collins, 1969; Moran, Fehn, and Ray, 1998). Since crude oil itself has much lower concentration of about 10–100 ppb of iodine (Fehn and others, 1987; Tullai, Tubbs, and Fehn, 1987) the elevated concentration results from the release of iodine from organic

material that had accumulated in the subsiding basin and the subsequent concentration by dewatering and compaction of sediments (Collins, 1969; Moran, 1996). Micropaleontological examinations have revealed the algal strands in iodine rich rocks (Collins, Bennett, and Manuel, 1971). Harrison and Summa (1991) calculated the vertical velocities of the fluids in sedimentary basins and suggested the release of iodine from the thermal alteration of organic matter into the solution. Synder and Fabryka-Martin (2007) reported the formation water to have  $^{129}\text{I}/\text{I}$  ages, after accounting for contribution from in situ production, to be similar to the age of source of organic matter with which the water is, or has been in contact. Moran, Fehn, and Hanor (1995) inferred, after studying the source age and fluid movements in sedimentary basins, about long range vertical migration for the iodine, the brines and the associated hydrocarbons.

Another possibility of a surface reaction between the migrating hydrocarbons and the atmospherically derived molecular iodine in presence of light has been suggested by few workers (Allexan and others, 1986; Tedesco, 1995; Leaver and Thomasson, 2002; Goudge, 2009). The insoluble iodo-organics are formed, which collect near the surface and act as one of the natural integrator of hydrocarbon seepage. Soil iodine anomalies and hydrocarbon accumulations are shown to be strongly correlated in the results of Leaver and Thomasson (2002) where case studies of soil iodine geochemistry have been related to subsequent drilling results. A number of studies have reported the photo-reduction of molecular iodine by organic matter (Spokes and Liss, 1996; Truesdale, 2007). Photochemically induced free radical chain reactions of soil iodine with the seeping hydrocarbons have been proposed where in near infrared region, the iodine radicals react with the hydrocarbons to produce iodo-alkyl compounds like ethyl iodide, propyl iodides, etc. (Tedesco and others, 1987; Goudge, 2009). Through atomization and evaporation, atmospheric iodine is delivered from the oceans to the atmosphere and then to soils by precipitation (Vinogradov, 1959). The physico-chemical properties of iodo-organic compounds such as stability, water insolubility and dependence on organic matter indicate toward the possibility of a covalently bonded iodorganic as primary form of iodine in soil overlying hydrocarbon accumulations (Allexan and others, 1986; Tedesco and others, 1987; Tedesco, 1995; Goudge, 2009).

The low volatility and high molecular weight of iodo-organic compounds make the transport of these compounds from subsurface depths improbable. The possibility of production of large amount of iodo-organics in the subsurface is small, as oil contains little iodine while the surrounding brines contain high concentration of iodine as sodium or potassium salt which are relatively inert to organics (Tedesco and others, 1987). In comparison to other halogens, the low bond dissociation energy of iodine of  $\sim 35$  kilocalories per mole of quanta, high wavelength of maximum response ( $\lambda_{\max} = 2580$ ) and molar extinction coefficient ( $\epsilon_{\max} = 378$ ) facilitate the formation of iodide radicals in the near infra-red region at a wavelength of  $7800 \text{ \AA}$ . Thus, low energy electronic transitions and availability of unsaturated, aromatic hydrocarbons, occurring in soil from natural seepage of petroleum, cause favourable conditions for iodine to form covalent bonds with hydrocarbons (Allexan and others, 1986; Tedesco, 1995). The other possible sources of iodine have been suggested to be low because of the fact that all rock forming minerals and evaporites commonly identified as the sources of brine constituents have extremely low concentration of iodine ( $<10$  ppb, Dean and Schreiber, 1987; Moran, Fehn, and Hanor, 1995) and possible sinks for iodide are limited because of its relatively large ionic radius inhibiting surface adsorption or substitution into common rock forming minerals. In the present work, the authors have reported the varied iodine concentration (1.1–19.3 ppm) that is found in the surface soils which may be overlying oil and gas seepage area, as supported by other geoscientific data. However, the chemical form in which iodine is present in the soil has not been taken as the part of this study.

## GEOLOGIC SETTINGS AND HYDROCARBON PROSPECTS

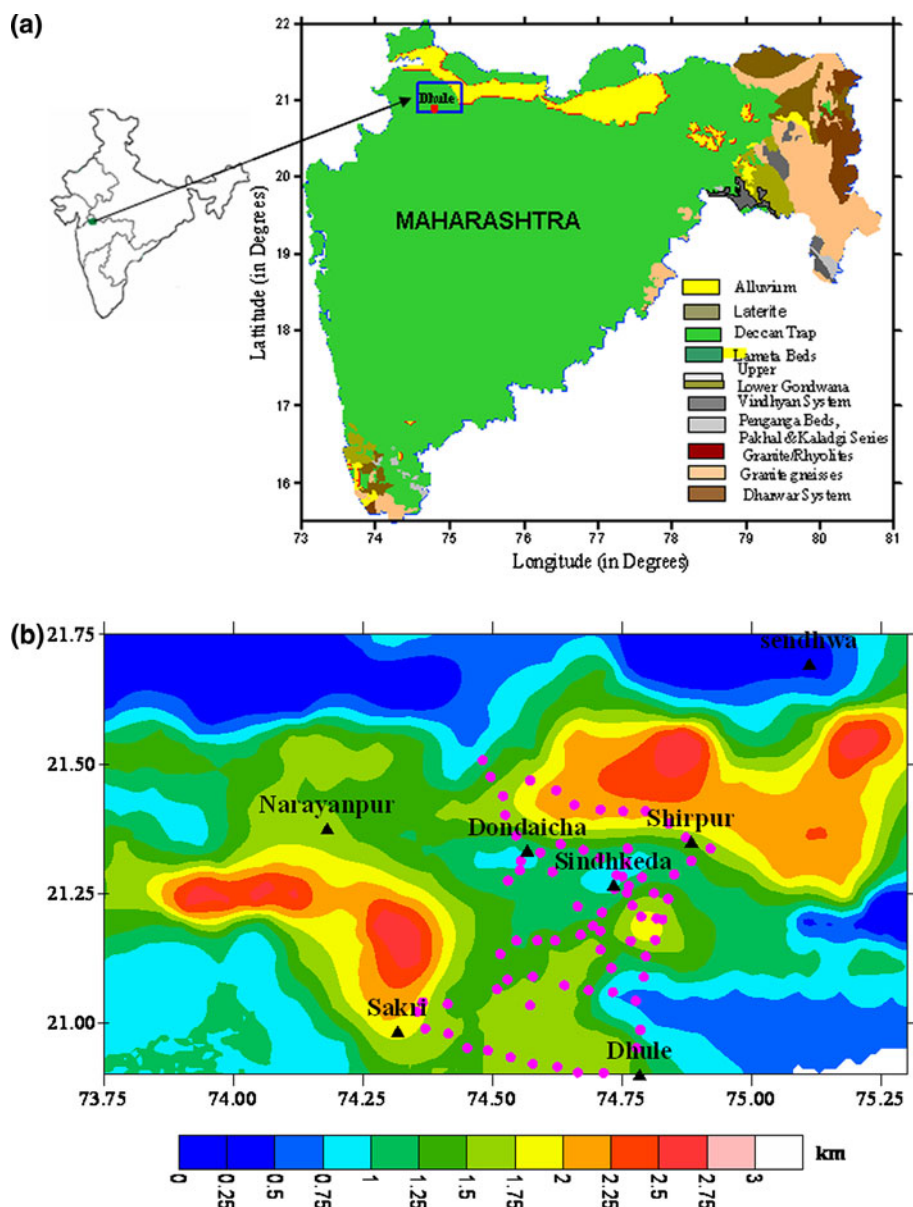
The Deccan Syncline is an intracratonic basin, covering an area of  $273 \times 10^3 \text{ km}^2$  in the western and south central part of India. Most of the basin is covered under the lava flows of the Deccan basalts of late Cretaceous, below which substantial thickness of Gondwana–Mesozoic sediments exist (Infraline, 2002). The area undertaken for this study is in and around the Dhule district of northern Maharashtra (Fig. 1a). The tectonic map of Deccan Syncline showing some of the major structural features, updip, pinchouts, and fault closures is

illustrated in Figure 2. The basin is bounded to the north by the Narmada–Son rift and to the south and east by Precambrian sediments and Archaean metamorphic rock exposures. The western limit is defined by longitudinal fissures through which there was eruption of basaltic lavas. The Kaladgi and Bhima depressions and Pranhita–Godavari graben, both with exposures of Gondwana–Mesozoic sediments lie on its southern and eastern margins, respectively, and the Narmada graben with Mesozoic sediments lies on its northern side (DGH, 2006). The study area is demarcated on the geological map of Maharashtra in Figure 1a, and the detailed sample location is shown over the sediment thickness map in Figure 1b.

The generalized stratigraphy of the Deccan Syncline is illustrated in Table 1. The biotite gneisses form the basement. Sediment deposition in Proterozoic period was followed by a long period of quiescence after which the glacial to fluvio-glacial sediments of Gondwana Group were deposited during the Carboniferous period. Over the exposed surface of the Gondwana rocks and older Archaean metamorphics, depositions of lacustrine and fluvial sediments known as Lameta beds occurred. Marine equivalents of the Lameta beds are the Bagh beds of the Narmada Valley, which are exposed in isolated outcrops in the Dhule district. These are basically conglomerate, overlain by sandstone and shale beds, and nodular, argillaceous limestone upward (Deshpande, 1998). The thickness of lower and upper Gondwana sediments along with Lameta beds is  $\sim 2$ – $3.5$  km (DGH, 2006; Harinarayana, 2008). The voluminous outpouring of the lava during the Late Cretaceous was one of the major events, giving rise to the famous Deccan volcanic province of India.

Large dyke swarm, trending roughly ENE–WSW and parallel to the graben of the Narmada and Tapi rivers is seen to occur in the Narmada–Tapi region of Northern Maharashtra (Auden, 1949; Deshmukh and Sehgal, 1988; Ray, Sheth, and Mallik, 2007). Major portion of Maharashtra is covered with black soil, which is the weathered product of the basalts. Laterite and alluvium with more than 200-m thickness has been observed at number of places (Deshpande, 1998). The geophysical studies have shown the presence of  $\sim 2.5$ -km thick Mesozoic sediments concealed below 2-km-thick Deccan Trap (DGH, 2006; Rao and Reddy, 2005; Harinarayana, 2008). The map of the sediment thickness modified after DGH, 2006 is shown in Figure 1b.





**Figure 1.** (a) Geological map of Maharashtra showing the study area (after Deshpande, 1998). (b) Detailed sample locations plotted on the sediment thickness map of Deccan Syncline (after DGH, 2006).

The Deccan Syncline is category IV sedimentary basin of India, which, on analogy with similar hydrocarbon-producing basins in the world is deemed to be prospective with uncertain potential (DGH, 2007). The sandstones of Mesozoic Sequence and Gondwana Group are the likely petroleum productive zones in the basin. The probable source rocks are the dark gray shales and marls of lower Gondwana and the caprocks might be the intraformational shales and the Trap flows (DGH, 2006).

High geothermal gradient (Biswas, 1982; Biswas and Deshpande, 1983; Zutshi, 1991; Pandey and Agrawal, 2000) and the tectonic history of the basin indicate the thermal subsidence and burial (Gomos Jr., Powell, and Norton, 1995; Schutter, 2003; Rohrman, 2007) to be significant enough to cause the maturation of organic-rich sediments and, most likely, the generation and expulsion of hydrocarbons. The Deccan Trap Volcanics have preserved the underlying Mesozoic strata and their hydrocarbons

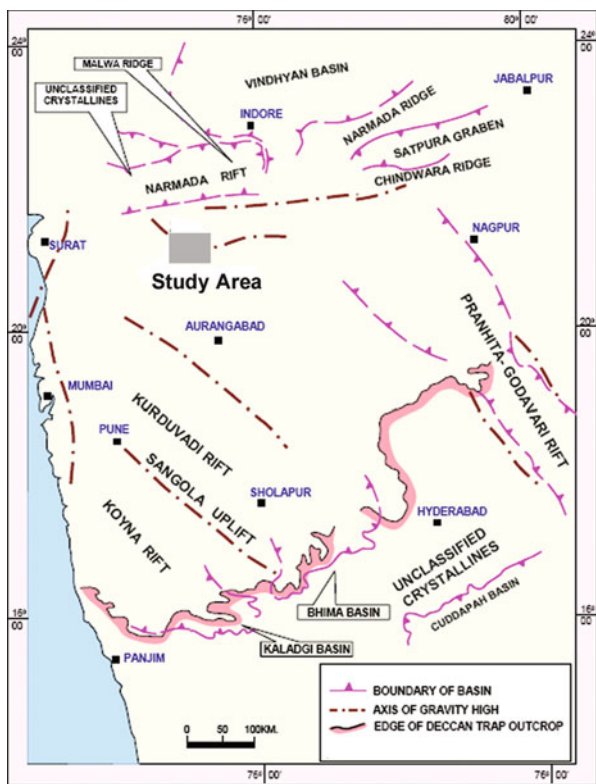


Figure 2. Tectonic map of study area showing the major structural features (after DGH, 2006).

from exposure and erosion over a long period of time from Cretaceous to the present day (Zutshi, 1991).

The area adjoining Nandurbar, beneath Narmada–Tapi region of Deccan Syncline, has been reported to show the presence of light gaseous hydrocarbons, methane (C<sub>1</sub>), and the sum of ethane, propane and butane (ΣC<sub>2+</sub>) in the range of 3–1187 and 1–1449 ppb, respectively, and the carbon isotopic signatures of these hydrocarbons show the thermogenic origin (δC<sup>13</sup> = CH<sub>4</sub> in the range from –24 to –39.4‰ PDB (Vardhan and others, 2008). Gas has been struck in the western part of the Krishna–Godavari basin at Mandapeta well in Chintalpudi Sandstone of Upper Gondwana, which is an encouraging factor for exploration in Gondwana sediments (DGH, 2006).

### SAMPLING AND ANALYTICAL PROCEDURES

#### Sampling

High concentration of thermogenic hydrocarbons (C<sub>1</sub>–C<sub>5</sub>) around Nandurbar, northwest of Dhule, has been reported by Vardhan and others

Table 1. Generalized Stratigraphy of the Deccan Syncline Basin (After Deshpande, 1998)

Age Range	Formation/ Group	Anticipated Thickness (km)	Lithology	Geographic Distribution
Recent– Pliocene	Alluvium Laterite, sand			Nagpur, Bhandara , Candrapur, Wardha Jalgaon, Kolhapur Satara Thane, etc.
Early Paleocene –Cretaceous	Deccan Traps	1–2	Basalts	Most of the state from west of Nagpur to Arabian sea coast
-----Unconformity-----				
Late Cretaceous	Lameta, bagh beds	2–3.5	Arenaceous limestone, Sandstone shale	Nagpur, Candrapur Dhule, Gadchiroli Yavatmal
Middle Triassic	Upper Gondwana			
-----Unconformity-----				
Triassic– Carboniferous	Lower Gondwana			Nagpur, Chandrapur Yavatmal
Proterozoic	Penganga beds			Gadchiroli, Ratnagiri
Archean	Limetsones, shales Sausar, Sakoli, Amagaon, Unclassified Gneisses			Nagpur, Bhandara Ratnagiri, Sindhudurg

(2008). This study is carried out in the eastern side of Nandurbar to investigate the microseepage of light hydrocarbons through the distribution of soil iodine concentration. The samples have been collected in and around the Dhule district, northern Maharashtra. The reconnaissance survey along the existing roads can help one to evaluate a large tract of land for its hydrocarbon resource potential and prioritize the areas for further exploratory studies. The soil samples have been collected from along the existing roads, nearly 40–50 ft away, avoiding the possible anthropogenic contaminations. Surface sampling is recommended to get the most repeatable values for the iodine as the soil and the related soil heterogeneity is less uncertain at surface compared to depth (Tedesco and others, 1987; Goudge, 2007). The detailed sample collection points are shown on the sediment thickness map of the Deccan Syncline basin in Figure 1b. A total of 72 samples were collected from a depth of top 2–6 in. of the soil, at an interval of 5 km using a shovel. The samples were sealed in the resealable plastic bags with their sample number and Global Positioning System (GPS) locations marked. Disturbed or excavated areas, soils contaminated with hydrocarbons, chemicals or animal wastes, swamps and areas under water shed were avoided for sampling. While collecting the samples, rocks, coarse materials, plant residues, and animal debris have been excluded.

### Iodine Measurement

A 100 mg of <math>63\ \mu\text{m}</math> size soil sample was treated with 5 mL of 10% Tetra methyl ammonium hydroxide in savillex pressure decomposition vessel at  $80^\circ\text{C}$  for 6 h. After cooling, the solution was diluted with Millipore water, and 1 mL of 250 ppb antimony was added to act as internal standard and the volume was made up to 25 mL. The final solution containing 2% TMAH and 10 ppb antimony was centrifuged at 3000 rpm for 20 min and filtered through Wattman No. 1 filter paper to get the clean solution. The analyses of samples, blank, and soil standards were performed using Perkin-Elmer Sciex DRC II Inductively Coupled Plasma Mass Spectrometer (ICP-MS). A matrix matching international soil reference material SO-1 (I = 12 ppm) was used to calibrate the ICP-MS. To evaluate the accuracy of the methodology, two different dilutions of soil reference material; SO-1 (25 and 50 mg), SO-3, and SO-4 were analyzed as

unknown samples and compared with the certified values. Appendix 1 provides the details of these soil reference materials, where the measured values are in close agreement with the certified values. The relative standard deviation (RSD) as three times the standard deviation of the blank is  $<1\%$ , and the limit of detection is 0.027 mg/kg. The details of the sample preparation and instrument parameters are mentioned elsewhere (Balaram and Rao, 2003; Mani and others, 2007).

### Total Organic Carbon (TOC) Measurement

About 1.5 g of  $63\ \mu\text{m}$  soil sample was treated with 3–4 drops of HCl to remove the inorganic carbon and kept overnight at  $50^\circ\text{C}$  in oven. About 50 mg of the dried, HCl-treated sample was loaded onto the Quartz boat and transferred to the furnace of the Solid Module  $1000^\circ\text{C}$ , a modification of the Lique Total Organic Carbon analyzer basic unit (Elementar Analysensysteme GmbH) for total organic carbon measurement. Similar procedure was adopted for the samples, blank, and the soil standard (Boden Soil Standard, 4.1% TOC). The  $\text{CO}_2$  released due to the chemical oxidation of the organic carbon is measured by the Infra Red (IR) detector and expressed in wt%. The %RSD of the procedure is  $\leq 1\%$ .

## RESULTS

This study conducted in the part of Deccan Syncline shows iodine concentration varying from 1.1 to 19.3 ppm. The %RSD of the selected samples has been determined, and the values obtained are  $\leq 1$ . The TOC measured for another aliquot of the soil samples range from 0.1 to 1.3%. Appendix 2 provides the concentration of iodine along with the RSD and the TOC content in part per million (ppm).

In general, if the samples display a near normal distribution, then the mean represents the background, and samples with one or more standard deviations are considered anomalous (Tedesco and others, 1987; Abrams, 2005). The frequency distribution pattern of iodine, differentiating between the background and the anomaly, based on mean and standard deviation (mean:  $5.1 \pm 2.7$  ppm) is shown in Figure 3. The histogram (class interval = one standard deviation) shows a Gaussian nature with skewness toward the right, indicating the presence of

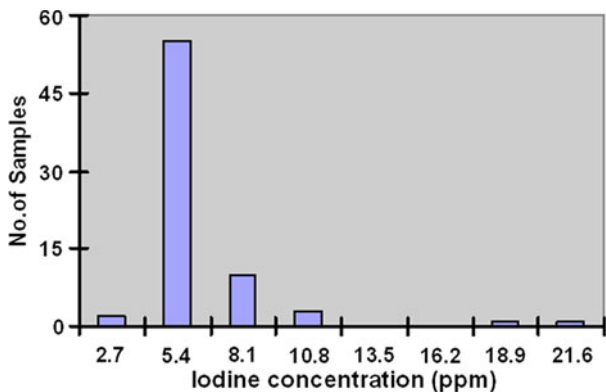


Figure 3. Histogram for soil iodine concentration.

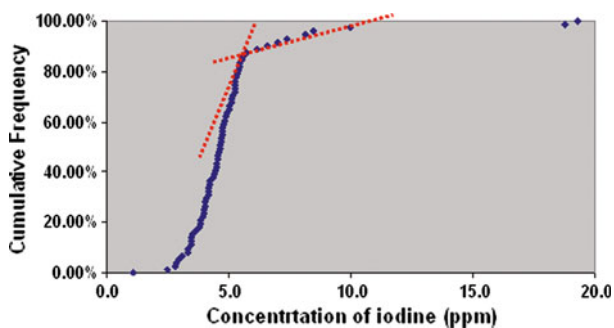


Figure 4. Probability plot for soil iodine concentration.

anomalous populations. The soil iodine concentration plotted against the cumulative percentage is shown in the probability plot (Fig. 4). The cumulative frequency plot displays the Gaussian distribution as straight line. The change in slope of the line shows the departure from the normal or background population, and the anomalous distribution can be recognized with the upward deflection of the curve. From the frequency graph (Fig. 4), a population of <5 ppm is observed, which has been defined as the background and used further to classify the iodine concentration into three groups, <5 ppm, 5–8 ppm, and >8 ppm, based on mean and standard deviation. Defining the threshold between the anomaly and background depends largely on the dataset. In this study, a lower value of background has been defined, as this preliminary reconnaissance study is carried out on a frontier basin for initial screening of priority locations for the probable hydrocarbon prospective areas and on relatively less number of samples. About 35% of the data has iodine values between 5 and 8 ppm, and a small percent (7%) of higher values of iodine (8–19.3 ppm) shows skewness in the positive direction.

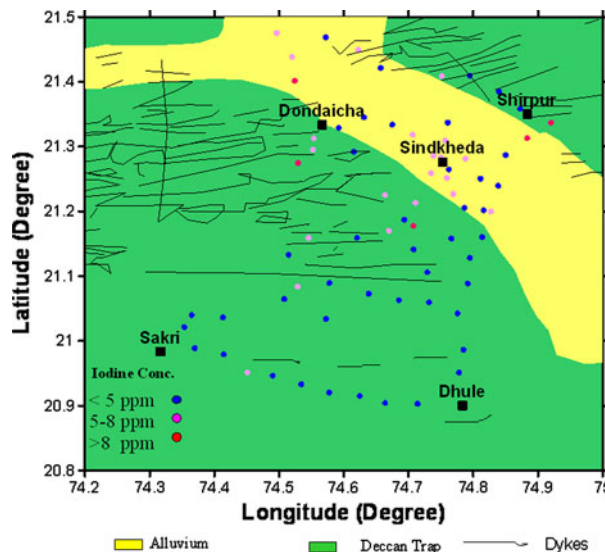


Figure 5. Iodine concentration distributions shown over the exposed Dyke swarm (after Ray, Sheth, and Mallik, 2007) in Dhule–Nandurbar area of Deccan Syncline.

DISCUSSION

The concentration distribution of iodine in surface soils of Dhule area is represented in Figure 5. It is observed that high iodine concentration occurs mainly over the northern part of the sampled region, especially in and around the exposed alluvium overlying Deccan Trap. Elevated concentrations of iodine between 5 and 19.3 ppm are found in areas over and adjoining Dondaicha, Sindkheda, and Shirpur. A few scattered points are seen lying around Sakri and north-west of Dhule. When iodine concentrations are plotted on sediment thickness map of Deccan Syncline, the samples overlie the sedimentary thickness ranging between 0.75 and 2.75 km. As the Gondwana sediments are expected to form the main source and reservoir facies (DGH, 2006), investigations in the areas of thick sedimentary rocks of Deccan Syncline may be helpful to prioritize the regions for further activities. The northern part of sampled area, which reflects a majority of high iodine values, has sediment thickness of about 2–2.25 km (Fig. 1b). The middle region or the basement high—separating two relatively thick sediment sections i.e., north of Shirpur and north of Sakri—shows the elevated concentration of iodine (Fig. 1b). The southern part of the study region does not show the basement high and has comparatively thin sedimentation with lower values of iodine (<5 ppm). The iodine anomalies are associated with the fixing of iodine in soils because of the



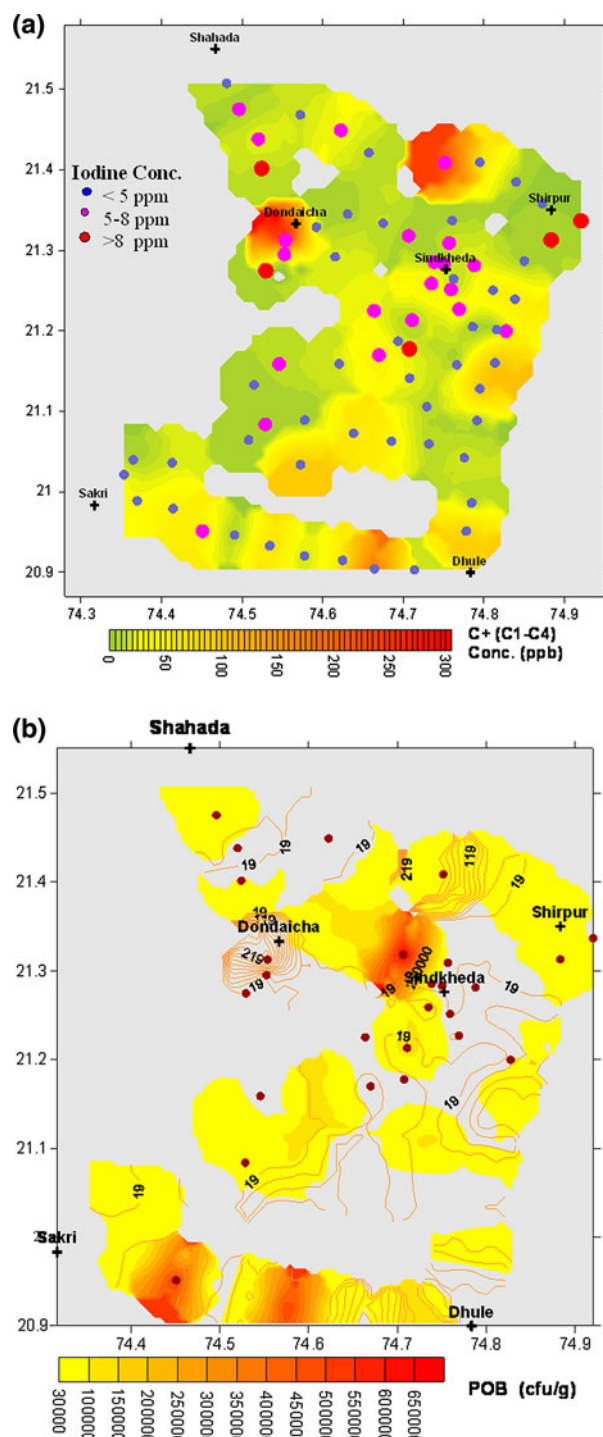
interaction with the seeping hydrocarbons, thus a more likely inference drawn from this concentration distribution is that no migrating oil accumulations are present in the southern part which is showing the background levels of iodine.

The major iodine highs are associated with ENE trend which is coincident with the occurrence of dyke swarms in the area (Fig. 5). Focused fluid migration along faults, discontinuities, or unconformities is much more effective than non-focused seepage through a sedimentary column (Abrams, 1992; Brown, 2000). Intrusions, such as sill and dykes, represent a preferential pathway for fluids and show active hydrocarbon seepage (Duranti and Mazzini, 2005; Gay and others, 2006). Reactivation of dyke-filled fracture systems and hydrocarbon migration-seepage pathways appear to be controlled by the reactivation of the pre-existing fractures and dykes within the basement (Rollet and others, 2006). The dyke swarm occurring in Narmada-Tapi region may provide preferential migratory path for hydrocarbon microseepage. Linear tholeiitic dykes, extending over 14500 km<sup>2</sup> in the Nandurbar-Dhule area and dioritic gabbros up to 3 m have been found exposed near Shirpur and Shahada regions of Deccan Syneclise. The Shahada and Shirpur gabbros lie along ENE-WSW structural ridges, which reflect a prominent long fault, greater than 30 km in length, coinciding with the Narmada-Satpura-Tapi regional tectonic trend (Sheth and others, 1997). The iodine values between 5 and 19.3 ppm are observed in the northern part of the area where the dykes are prominently exposed (Fig. 5). Owing to the Tertiary and Quaternary alluvium along Tapi river (>30 km wide and 200-400 m thick) which caps the basalt section, the continuity of the dykes is unknown (Sheth and others, 1997) and the mapping of dykes has not been possible. The seepage of light hydrocarbons toward the shallow surface might assist the fixing of iodine in soil, causing high concentration of iodine near Dondaicha, Sindkheda, and Shirpur. The southern part has iodine values less than 5 ppm and is associated with relatively fewer and widely spaced dykes adjoining Dhule and Sakri. The pattern of iodine concentration distribution follows the ENE trend which is supported by the emplacement of dykes in the region.

A direct indicator of seepage is the light gaseous hydrocarbon, methane through butane, adsorbed onto the soils. The hydrocarbon gases are assumed to be migrating from the subsurface oil and gas reservoirs and provide direct evidence of microseepage (Jones and Drozd, 1983; Schumacher, 1996; Abrams,

2005; Mani, 2008). The light gaseous methane through butane ( $C_1-C_4$ ;  $\sum C_{1-4}$ ), desorbed from the soil samples of Deccan Syneclise are contoured based on mean in Figure 6a (modified after Kumar, 2009). The higher concentrations (~80-300 ppm) of these hydrocarbons are seen around Dondaicha, Sindkheda, north-west of Shirpur, and to some extent around west of Dhule. The iodine concentrations, posted over the light hydrocarbons ( $\sum C_{1-4}$ ), show the iodine highs (>5 ppm) overlying the high concentration of soil gases near Dondaicha, Sindkheda, and north-west of Shirpur (Fig. 6a). The iodine lows are observed north east of Sakri and north of Dhule and between Dondaicha and Shirpur, where the soil gases are also found in low concentration (<60 ppm; Kumar, 2009). However, south of Shahada and Shirpur and area west of Dhule show couple of iodine values which do not appear to correlate with light hydrocarbons except for two to three scattered points. The variations existing in the use of different surface geochemical indicators are governed by the extreme complexities of processes controlling them. Factors such as supply of hydrocarbons from subsurface, microbial activity, etc., influence the distribution of the variables in the soil. Overcoming all of these problems is practically impossible; however, if the soil gas survey suffices the condition that the gases are liberated in proportion to the amounts present and that the analytical results bear some relationship to one another and also to other investigative parameter, like the soil iodine here, thus, it allows the identification of potentially prospective areas (Ruan and Fei, 2000). Observations with still larger number of sample points around the area can account for the weak correlation of iodine with soil gas in this region. In general, the pattern of iodine values closely associates itself with the light hydrocarbon gases. Occurrence of dyke swarms in the northern part around Dondaicha, Sindkheda, and Shirpur might assist the light gaseous hydrocarbons to reach the near-surface soils, thereby becoming available for binding with the soil iodine. The southern part of the study area has the fewer and widely spaced dykes (Fig. 5), suggesting the possible control of these structural features in microseepage toward the shallow surface.

Another widely used indirect indicator for the near-surface oil and gas prospecting is hydrocarbon-oxidising microbial populations which exclusively utilize the seeping hydrocarbons as their carbon nutrient source for metabolic activity and growth (Wagner, Piske, and Smit, 2002). The bacteria are



**Figure 6.** (a) Soil iodine distribution shown over the contoured light gaseous hydrocarbons (C<sub>1</sub>-C<sub>4</sub>;  $\sum C_{1-4}$ ) (after Kumar, 2009). (b) Composite map of propane oxidizing bacteria and soil iodine (after Rasheed and others, 2008). The iodine concentrations above 5 ppm are posted in red dots while the light hydrocarbon concentration ( $\sum C_{1-4}$ ) is shown in red contours (after Kumar, 2009).

found mostly enriched in the shallow soils/sediments above hydrocarbon-bearing structures and can differentiate between hydrocarbon-prospective and non-prospective areas (Miller, 1976). The bacterial counts range between  $10^3$  and  $10^6$  cfu/g (colony-forming units/gram) in soils receiving hydrocarbon microseepage, depending on ecological conditions (Hanson and Hanson, 1996; Wagner, Piske, and Smit, 2002). The propane-oxidizing bacterial count in the soil of the studied area of Deccan Syncline has been observed to be in the range of  $1.0 \times 10^2$  to  $6.7 \times 10^5$  (Rasheed and others, 2008). The iodine concentrations >5 ppm have been plotted over the propane-oxidizing bacteria (POB) count map (Fig. 6b). A good correlation of the two indirect indicators is seen in the northern part of the sampled area around Sindkheda, Dondaicha, and Shirpur. In the southern part, the area east of Sakri also has high iodine value overlying the POB. A few spotted points are seen scattered around south of Shahada and Sindkheda, and west of Dhule where a perfect match of the two indicators is not observed, but the overall agreement between the two methods reflects the iodine and microbial correlation in the northern region of the study area.

The iodine highs over the oil and gas fields have largely been attributed to the association of iodine with the organic matter. If the iodine binds with the surficial organic matter contributed mainly from the plant and bacterial organic matter, rather than the seeping hydrocarbons, then false anomalies can mislead the interpretations. Samples showing high values of surficial TOC content may show high values of iodine. To rule out such a possibility, correlation of surficial TOC was seen with the soil iodine concentration. The TOC varies between 0.1 and 1.3%, and correlates poorly with the soil iodine (correlation coefficient,  $r = 0.01$ ). This suggests that iodine is not bound to the surface organic carbon and indicates toward the possibility of migrated hydrocarbons as source for fixing the soil iodine. The lack of binding of iodine with organic matter is also supported by the soil type that exists in the study area. Soils generally contain iodine in the concentration range of 0.01–6 mg/kg (Kebata-Pendias and Pendias, 1984). The average highs of iodine in continental shales and limestone are 1.8 and 2.5 ppm, respectively, whereas graywacks and sandstone on an average contain 150 and 120 ppb, respectively (Muramastu and Wedepohl, 1998). The Bagh and Lameta beds occurring below the Traps show such lithology

(Table 1) and are seen in the isolated outcrops in Dhule district (Deshpande, 1998). Major portion of the study area is covered with black soil, which is the weathered product of the Basalts. At places, laterite and alluvium with thicknesses ranging from 200 to 400 m are present. Black soils are highly argillaceous, very fine-grained and dark, and contain a high proportion of calcium and magnesium carbonates; however, they are poor in organic matter. Lithologies of such types have low iodine-fixation potential (Fuge, 1987). The sampled region mostly consists of alluvium and black soil, and the geometric mean value ( $\mu\text{g/g}$ ) of bound iodine in these fractions of soil is  $<2.2$  (Fuge and Johnson, 1986). The iodine values reported here are significantly higher than the average distribution values of iodine in such type of soils. This observation supports the possibility hydrocarbon components migrating from subsurface as source for interaction with soil iodine. Integration of iodine with the light hydrocarbons and microbial populations reveal the northern part adjoining Dondaicha, Sindkheda, and Shirpur to have anomalous values of these indicators (Fig. 6b). The southern part, especially east of Sakri, shows decreasing concentration of iodine, light hydrocarbons, and POB.

## CONCLUSION

The soil iodine distribution in parts of the Deccan Syneclise has been studied to understand its association with the microseepage and infer about the hydrocarbon prospects of the basin. The iodine concentration ranges from 1.1 ppm to 19.3 ppm and is distributed over the entire study area, differentiating the background and the anomalous regions. The northern part of the sampled region shows significant iodine concentrations in range of 5–19.3 ppm especially near Dondaicha, Sindkheda, and Shirpur. The results of the iodine studies closely associate with the corresponding direct and indirect surface geochemical indicators of hydrocarbon microseepage, the adsorbed light gaseous hydrocarbons, methane through butane, and the propane-oxidizing bacterial populations in the soil. The soil types found in the area have low iodine-fixation potential, indicating negligible contribution toward the increase of iodine values. The TOC values correlate very poorly ( $r < 1$ ) with the iodine concentrations, suggesting the possibility of seeping hydrocarbons as source for the fixing of iodine in soils. The Mesozoic sedimentary thickness and emplacement of dyke swarms in Nandurbar–Dhule

area further substantiate these results. The pattern of iodine concentration distribution follows the ENE trend that is similar to the structural trend exhibited by the dykes, which may facilitate the seepage of hydrocarbons toward the shallow surface. Thus, the positive correlation observed among the geochemical, geological, and geophysical parameters in this study greatly increase the diagnostic value of near surface soil iodine as a predictive geochemical hydrocarbon tool in constraining the exploration risk for oil and gas. The results of this integrated study show that detailed investigations for locating the oil and gas reserves in the Mesozoic of the Deccan Syneclise basin can prove to be rewarding.

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## APPENDIX 1

Certified and Measured Concentrations of Iodine with %RSD Using ICP-MS in Soil Reference Materials

Reference Material Soil Standard	Certified Value (ppm)	Measured Value (ppm)	%RSD
SO-1 <sup>a</sup> (25 mg)	3	3 ± 0.00	0.00
SO-1 <sup>a</sup> (50 mg)	6	6.388 ± 0.69	0.83
SO-3 <sup>a</sup>	1	0.585 ± 0.32	0.58
SO-4 <sup>a</sup>	3	2.719 ± 0.17	0.59

<sup>a</sup>Replicates of six analyses

## APPENDIX 2

Iodine and TOC Concentration of Soil Samples (in ppm) from the Deccan Syneclise Basin

Sample ID	Iodine (ppm)	TOC (ppm)
DS/01/06	4.4	7132
DS/02/06	3.0	1641
DS/03/06	3.8	7443

continued

Sample ID	Iodine (ppm)	TOC (ppm)
DS/04/06	4.8	6975
DS/05/06	2.8	7230
DS/06/06	3.4	6743
DS/07/06	5.3	1477
DS/08/06	2.5	7519
DS/09/06	1.1	10000
DS/10/06	4.2	6681
DS/11/06 <sup>a</sup>	3.5 (0.22)	6944
DS/12/06	4.2	10860
DS/13/06	4.5	4366
DS/14/06	3.5	7962
DS/15/06	2.9	3343
DS/16/06	4.1	7394
DS/17/06	3.5	5163
DSS/18/06	3.5	6203
DS/19/06	4.1	8134
DS/20/06	5.3	6227
DS/21/06	4.2	5918
DS/22/06 <sup>a</sup>	4.7 (0.14)	4622
DS/23/06	4.8	5874
DS/24/06	5.4	6912
DS/25/06	5.0	7073
DS/26/06	3.7	5749
DS/27/06	4.5	6675
DS/28/06	4.9	5751
DS/29/06	5.3	6378
DS/30/06	4.2	13640
DS/31/06	4.0	6727
DS/32/06 <sup>a</sup>	4.8 (0.12)	6404
DS/33/06	4.0	4507
DS/34/06	5.2	6483
DS/35/06	5.3	6745
DS/36/06	5.6	5977
DS/37/06	4.7	6032
DS/38/06	8.2	8491
DS/39/06	18.8	8840
DS/40/06	4.6	6532
DS/41/06	4.7	8430
DS/42/06 <sup>a</sup>	4.6 (0.33)	6081
DS/43/06	5.3	8860
DS/45/06	4.9	6599
DS/46/06	5.1	5747
DS/47/06	4.8	7939
DS/48/06	4.7	7267
DS/49/06	5.5	8233
DS/50/06	7.0	7860
DS/51/06 <sup>a</sup>	8.5 (0.72)	5869
DS/53/06	6.6	8426
DS/54/06	19.3	5462
DS/55/06	5.0	7305
DS/56/06	3.1	6417
DS/57/06	3.4	6951
DS/58/06	4.0	9981
DS/59/06	3.9	6006
DS/60/06	4.2	8034
DS/61/06	5.4	6356
DS/62/06	4.9	6652
DS/63/06 <sup>a</sup>	6.2 (1.0)	5362

continued

Sample ID	Iodine (ppm)	TOC (ppm)
DS/65/06	4.5	6910
DS/66/06	5.7	5608
DS/67/06	10.0	5921
DS/68/06	4.6	7006
DS/69/06	3.9	6985
DS/70/06	3.8	7022
DS/71/06	5.6	7543
DS/72/06	5.4	6108
DS/73/06	7.4	7728
DS/74/06	5.1	6431
DS/75/06	4.4	5985

The RSD of iodine concentration in selected samples (<sup>a</sup>) is given in brackets

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