Surface Inorganic Iodine Speciation in the Indian and Southern Oceans from 12°N to 70°S

Running title: Iodine in the Indian Ocean

Rosie J. Chance[1*+], Liselotte Tinel[1+], Amit Sarkar[2‡], Alok K. Sinha[2], Anoop S. Mahajan[3], Racheal Chacko[2], P. Sabu[2], Rajdeep Roy[4], Tim D. Jickells[5], David Stevens[6], Martin Wadley[6], Lucy J. Carpenter[1]

1. Wolfson Atmospheric Chemistry Laboratories, Department of Chemistry, University of York, York, YO10 5DD, UK.
2. National Centre for Polar and Ocean Research, Ministry of Earth Sciences, Goa 403804, India.
3. Indian Institute of Tropical Meteorology, Ministry of Earth Sciences, Pune 411008, India.
4. National Remote Sensing centre (NRSC), Indian Space Research Organisation (ISRO), Department of Space, Government of India.
5. Centre for Ocean and Atmospheric Sciences, School of Environmental Sciences, University of East Anglia, Norwich Research Park, Norwich, NR4 7TJ, UK.
6. Centre for Ocean and Atmospheric Sciences, School of Mathematics, University of East Anglia, Norwich Research Park, Norwich, NR4 7TJ, UK.

+ Authors contributed equally to this manuscript

Current address:
‡ Ecosystem based management of marine resources, (EBMMR), Environment and Life Sciences Research Center, Kuwait Institute for Scientific Research, Salmiya, Kuwait

* Correspondence: Corresponding author rosie.chance@york.ac.uk

Key words: iodine, iodide, iodate, seawater, Indian Ocean, Southern Ocean

Contribution to the Field

The presence of iodide-iodine at the ocean surface impacts air quality and climate. Specifically, the reaction of iodide with ozone at the sea surface is the dominant source of iodine to the atmosphere, and a significant sink for tropospheric (pollutant) ozone. In the atmosphere, iodine takes part in further reactions which remove more ozone, affect greenhouse gas lifetimes and contribute to particle formation. The rate of ozone removal and iodine emission depends on the sea surface iodide concentration, but iodide measurements are sparse and have hitherto been almost completely lacking in the Indian Ocean basin. Here we present an extensive new set of sea-surface iodide measurements, spanning latitudes from ~12°N to ~70°S and including measurements from the Bay of Bengal, the Arabian Sea, the open Indian Ocean and the Indian Ocean sector of the Southern Ocean. This data set represents a substantial increase in the global coverage of sea surface iodide observations. It will help improve our understanding of marine iodine biogeochemistry, and our ability to predict the impact of iodine chemistry on the atmosphere. We compare the observed distribution to that in other ocean basins, and explore the drivers of the distribution using an ocean iodine cycling model.

52   **Abstract**
53
54   Marine iodine speciation has emerged as a potential tracer of primary productivity,
55   sedimentary inputs, and ocean oxygenation. The reaction of iodide with ozone at the sea
56   surface has also been identified as the largest deposition sink for tropospheric ozone and the
57   dominant source of iodine to the atmosphere. Accurate incorporation of these processes into
58   atmospheric models requires improved understanding of iodide concentrations at the air-sea
59   interface. Observations of sea surface iodide are relatively sparse and are particularly lacking
60   in the Indian Ocean basin. Here we examine 127 new sea surface (≤10 m depth) iodide and
61   iodate observations made during three cruises in the Indian Ocean and the Indian sector of the
62   Southern Ocean. The observations span latitudes from ~12°N to ~70°S, and include three
63   distinct hydrographic regimes: the South Indian subtropical gyre, the Southern Ocean and the
64   northern Indian Ocean including the southern Bay of Bengal. Concentrations and spatial
65   distribution of sea surface iodide follow the same general trends as in other ocean basins,
66   with iodide concentrations tending to decrease with increasing latitude (and decreasing sea
67   surface temperature). However, the gradient of this relationship was steeper in subtropical
68   waters of the Indian Ocean than in the Atlantic or Pacific, suggesting that it might not be
69   accurately represented by widely used parameterisations based on sea surface temperature.
70   This difference in gradients between basins may arise from differences in phytoplankton
71   community composition and/or iodide production rates. Iodide concentrations in the tropical
72   northern Indian Ocean were higher and more variable than elsewhere. Two extremely high
73   iodide concentrations (1241 and 949 nM) were encountered in the Bay of Bengal and are
74   thought to be associated with sedimentary inputs under low oxygen conditions. Excluding
75   these outliers, sea surface iodide concentrations ranged from 20 to 250 nM, with a median of
76   61 nM. Controls on sea surface iodide concentrations in the Indian Ocean were investigated
77   using a state-of-the-art iodine cycling model. Multiple interacting factors were found to drive
78   the iodide distribution. Dilution via vertical mixing and mixed layer depth shoaling are key
79   controls, and both also modulate the impact of biogeochemical iodide formation and loss
80   processes.

Introduction

Iodine is naturally present in the ocean, predominantly as the inorganic ions iodide (I^-) and iodate (IO_3^-). Iodine speciation is linked to many aspects of ocean biogeochemistry, and has been proposed as a tracer of primary productivity (Ducklow et al. 2018; Wong 2001), sedimentary inputs and oxygen status (Lu et al. 2018; Moriyasu et al. 2020). In addition, the concentration of iodide at the sea surface has recently attracted renewed interest from atmospheric chemists because of its impact on atmospheric composition and air quality e.g. (Sherwen et al. 2017; Cuevas et al. 2018).

The heterogeneous reaction of iodide with ozone at the sea surface has been identified as the largest, but also most uncertain, deposition sink for tropospheric ozone (Hardacre, Wild, and Emberson 2015), and the dominant source of volatile reactive iodine (as I_2 and HOI) to the lower atmosphere (Carpenter et al. 2013). Following emission from the ocean, reactive iodine species initiate catalytic ozone depletion cycles, and hence further influence the oxidative capacity of the atmosphere. Atmospheric iodine cycling results in the formation of iodide oxides, which have been implicated in the nucleation of particles in coastal marine areas (McFiggans et al. 2004; Allan et al. 2015).In remote marine locations, iodine chemistry may also indirectly contribute to the depletion of inorganic volatile species such as gaseous elemental mercury during the polar spring (Wang et al. 2014). The ozone-iodide reaction is now thought to be the dominant source of iodine to the atmosphere, with other sources (e.g. release of iodinated organic compounds by marine algae) contributing only around 20% of the total iodine flux to the atmosphere globally (Carpenter et al. 2013; Prados-Roman et al. 2015). To incorporate the sea surface ozone sink and/or iodine source into atmospheric models, iodide concentrations at the interface need to be predicted accurately. However, parameterizations for global sea surface iodide concentrations (MacDonald et al. 2014; Chance et al. 2014) have been limited by the relative scarcity of observations. This is particularly the case for the Indian Ocean basin, where only a few sea surface iodide observations have hitherto been reported (Chance et al. 2014), but atmospheric iodine chemistry has been investigated (e.g. (Mahajan et al. 2019).

In ocean waters, total inorganic iodine concentrations (the sum of iodide and iodate) behave approximately conservatively, with a value of around 450-500 nM across most of the oceans e.g. (Elderfield and Truesdale 1980; Truesdale, Bale, and Woodward 2000). Iodate is thermodynamically the more stable form under oxygenated conditions, and hence is present at higher concentrations throughout most ocean depths. However, at the ocean surface iodide concentrations are elevated and iodate depleted, despite this being thermodynamically unfavourable. Sea surface iodide concentrations typically range from undetectable to ~250 nM, with values higher than this only encountered as outliers (Chance et al. 2014). Iodide concentrations and the iodide to iodate ratio decline with depth below the euphotic zone, and iodide concentrations are generally very low (<20 nM) in oxygenated waters below ~500 m e.g.(Waite, Truesdale, and Olafsson 2006; Nakayama et al. 1989; Bluhm et al. 2011). Under low oxygen conditions, iodide becomes the thermodynamically favourable form, and is found to dominate in sub-oxic and anoxic waters (e.g. Cutter et al., 2018; Rue et al., 1997; Wong and Brewer, 1977). In addition to the two inorganic forms, iodine also occurs in association with dissolved organic matter as so-called dissolved organic iodine (DOI; e.g. (Wong and Cheng 1998)). In the open ocean, levels of DOI are typically low (<5% of the total iodine), but higher levels (e.g. 22% of total iodine) have sometimes been encountered (Wong and Cheng 1998). However, organic forms of iodine are abundant in estuarine environments, where they can represent up to 64% of the total iodine (Wong and Cheng 1998; Schwehr and

131 Santschi 2003). Deposition of particulate iodine to the ocean floor is only a small sink for
132 iodine, with most being remineralised in the water column (Wong et al., 1976). Meanwhile,
133 redox cycling of iodine in sediments is thought to occur (e.g. Price and Calvert, 1977) and
134 iodide-iodine may be released from reducing sediments (e.g. Cutter et al., 2018; Farrenkopf
135 and Luther 2002).
136
137 The ratio of iodide to iodate varies with location as well as depth. Sea surface iodide
138 concentrations exhibit a pronounced latitudinal gradient, with highest surface iodide
139 concentrations observed at low latitudes and in coastal waters (Chance et al. 2014). The
140 distribution of iodine species in the oceans is thought to result from a combination of
141 hydrodynamic and biogeochemical drivers, which are not yet fully understood (Chance et al.
142 2014). The formation of iodide from iodate in the euphotic zone is thought to be associated
143 with primary productivity, but the exact mechanism by which this occurs is not yet known
144 (Chance et al. 2010; Campos et al. 1996; Bluhm et al. 2010). Similarly, the route for iodide
145 oxidation back to iodate has been elusive, although recent work has suggested it may be
146 linked to bacterial nitrification (Zic, Caric, and Ciglenecki 2013; Hughes et al., 2020). The
147 lifetime of iodide with respect to oxidation is poorly constrained but thought to be relatively
148 long, with estimates ranging from several months (Campos et al. 1996; Zic, Caric, and
149 Ciglenecki 2013) or more (Hardisty et al. 2020), to as much as 40 years (Tsunogai 1971;
150 Edwards and Truesdale 1997). Given the relatively long lifetime of iodide in seawater, its
151 distribution is also strongly influenced by advection and vertical mixing e.g. (Campos et al.
152 1996; Truesdale, Bale, and Woodward 2000). Seasonal variations in biological activity and
153 ocean mixing may result in seasonality in iodine speciation in the mixed layer (e.g. Chance et
154 al., 2010). The interplay of these driving factors results in global scale correlations between
155 sea-surface iodide concentrations and sea-surface temperature (SST), and nitrate (Chance et
156 al. 2014), which have been used to predict sea-surface iodide fields (e.g. Ganzeveld et al.
157 2009; MacDonald et al. 2014; Sarwar et al. 2016). In particular, spatial variations in ocean
158 mixed layer depth are likely to be the primary cause of the widely used relationship between
159 sea surface iodide concentration and SST.
160
161 This manuscript explores an extensive new set of sea surface iodide and iodate observations
162 from the Indian Ocean and the Indian sector of the Southern Ocean, spanning latitudes from
163 ~12∘N to ~70∘S, regions which have been lacking in observations (Chance et al. 2014). To the
164 best of our knowledge, only three studies have previously examined iodine speciation in the
165 Indian Ocean, and all have been in the west of the basin (East African coastal area (Truesdale
166 1978) and Arabian Sea (Farrenkopf and Luther 2002; Farrenkopf et al. 1997)). Of the data
167 presented in these studies, there were only two sea surface iodide observations - both from
168 the Arabian Sea (see Figure 1) - that could be included in Chance et al., 2014. The aim of this
169 study was to substantially increase the number of observations in this region, in order to both
170 improve understanding of large-scale gradients in ocean iodine speciation, and to increase
171 global data coverage for model validation and the improvement of ocean iodide
172 parameterisations. All of these will contribute to the creation of more accurate boundary
173 conditions for atmospheric chemistry models.
174
175 **Materials and methods**
176
177 *Sample collection*
178
179   *[Figure 1]*
180

Samples were collected during three research cruises in the Indian Ocean and Indian sector of the Southern Ocean. Sampling locations for each cruise are shown in Figure 1. Samples were collected from the Bay of Bengal (BoB) during a zonal section cruise (Bay of Bengal Boundary Layer Experiment - BoBBLe) along 8°N, from 85.3°E to 89°E. The cruise took place between 24/6/2016 and 23/7/2016, on board the RV *Sindhu Sadhana*. In the Arabian Sea, samples were collected during the Rama mooring equatorial cruise IO1-16-SK (known as SK333 here), operated by Ministry of Earth Sciences, India (MoES) and National Oceanic and Atmospheric Administration, USA (NOAA), and taking place on the ORV *Sagar Kanya*. The cruise departed Chennai, India, on 23/08/2016 and returned to Sri Lanka on 23/09/2016. Samples were collected from the Southern Indian Ocean and Southern Ocean during the 9th Indian Southern Ocean Expedition (SOE9; Jan-March 2017), from Mauritius (22°S) to coastal waters of Prydz Bay, Antarctica (69°S) on board the MV *SA Agulhas*. Sampling included both (Antarctic) coastal and open ocean waters during this cruise.

During the BoBBLe and SOE9 cruises, surface water samples were obtained manually from the upper 30-70 cm of the sea surface using a metal bucket deployed over the windward side of the ship near the stern. Additional depth profile samples were obtained using a CTD (911 plus, Sea-Bird Electronics, USA) rosette equipped with 12 Niskin bottles. During the SOE9 cruise, depth profiles were taken at 17 CTD stations, and additional surface samples were taken (by bucket) at least twice a day along the entire cruise track (except when the ship was stationary for CTD stations). Sampling included two time-series, one at ~40°S, and one in coastal Antarctic waters at ~68°S (around the Polar Front), during which samples were collected at 4 or 6 hour intervals for up to 72 hours. During the SK333 cruise, samples were only collected using a CTD rosette. Sample dates, times and locations for all cruises are given in the online dataset available from the British Oceanographic Data Centre (http://doi.org/czhx). In this manuscript, both shallow CTD samples (depth ≤ 10 m) and 'bucket' samples will be considered to be comparable, and representative of the ocean surface. This follows the approach taken in previous examinations of sea surface iodide concentrations (Chance et al. 2014). Note only surface samples are considered in this manuscript, although selected depth profiles are presented in the Supplementary information to aid interpretation of surface concentrations.

Immediately following collection, samples were filtered (Whatman GF/F) under gentle vacuum, and transferred to 50 mL polypropylene screw cap tubes. Duplicate aliquots were prepared for each sample. Aliquots were either stored at 4°C for on-board iodide determination within 24 hours, or frozen at -20°C for transport back to our laboratories for analysis. The majority (91%) of frozen samples were analysed within 12 months of collection, and all analyses were complete within 17 months of collection. Inorganic iodine speciation is preserved in frozen samples (≤-16°C) for at least one year (Campos 1997). Repeat analyses of high and low concentration samples indicated that iodine speciation was preserved during the storage period. To avoid possible contamination, sample bottles for the iodine samples were kept strictly separated from the dissolved oxygen reagents containing iodine.

Iodide and iodate analysis
Iodide was determined by cathodic stripping square wave voltammetry (Campos 1997; Luther, Swartz, and Ullman 1988) using a µAutolab III potentiostat connected to a 665VA stand (Metrohm) with a hanging mercury drop electrode, an Ag/AgCl reference electrode and a carbon or platinum auxiliary electrode. 12 (or 15) mL of the sample was introduced to a glass cell and 90 (or 112) µL of Triton X-100 (0.2%) was added. The sample was purged

with N_2(oxygen free grade) for 5 minutes before each measurement. The deposition potential was set at 0 V and deposition times were typically 60 s; scans ranged from 0 to -0.7 V, with a step of 2 mV, a 75 Hz frequency and a 0.02 V wave amplitude. Each scan was repeated 5-6 times, with scan repeatability equal or better than 5%. Calibration was by 2 or 3 standard additions of a KI solution (~10^{-5} or 10^{-6} M). Precision was estimated by repeat analysis (n = 6) of selected seawater samples over period of ten days and was found to be lower than 7% relative standard deviation.

Iodate was measured using a spectrophotometer (UV-1800, Shimadzu; 4 decimal places) after reduction to iodonium (I_3^-) (Jickells, Boyd, and Knap 1988; Truesdale and Spencer 1974). 2.3 mL of the sample was introduced in the 1 cm UV quartz cell, 50uL of sulfamic acid (1.5 M) was added, and the first absorbance value was obtained after 1 minute. Then 150 μL of KI (0.6 M) was added and the second absorbance read after 2.5 minutes. Iodate concentrations were calculated from the difference between the two absorbances. Calibration was performed daily using a series of KIO_3 standard solutions. Samples were measured at least in triplicate with repeatability better than 5%; reported values are means. Reported errors are calculated by propagation of the standard deviation on the repeated measurements, the errors on the fit of the calibration and error on the volumes pipetted. Note that strictly, this method measures all inorganic iodine in oxidation states from 0 to +5, but as this fraction is dominated by iodate it is taken as a measure of iodate.

Supporting measurements
Samples from CTD stations use the temperature, salinity and depth data directly obtained from the CTD. Precision of these measurements was as follows: temperature: ± 0.001 °C; conductivity: ± 0.0001 S m^{-1}; depth: ± 0.005% of the full scale. CTD salinity was calibrated using a high-precision salinometer (Guildline AUTOSAL). Temperature and salinity of manually collected 'bucket' samples were determined using an outboard thermometer, and the salinometer, respectively.

Samples for nitrate (NO_3^-) analysis were collected in 250 mL narrow mouth polypropylene amber bottles (Nalgene). Each bottle was rinsed twice with the sample water prior to collection. Analysis was performed onboard as soon as possible after sample collection, using an SKALAR SAN+ segmented continuous flow autoanalyzer. Precision and accuracy of NO_3^- measurements were ±0.06 and ±0.07 μM, respectively.

Ocean Modelling
The ocean iodine cycling model described in Wadley et al. (Wadley et al. 2020) was used to evaluate which physical and biogeochemical processes drive the observed trends in iodide concentration in the Indian Ocean and Indian sector of the Southern Ocean. The model comprises a biogeochemical model of iodine cycling embedded in a three-dimensional global ocean circulation framework, and has been calibrated using the data from a recently available extended global sea surface iodide compilation (Chance et al. 2019) which includes the current data set, plus additional depth resolved iodide measurements (see (Wadley et al. 2020) for details). In the model, iodide production is driven by primary production, and iodide loss (by oxidation) is linked to biological nitrification. A spatially variable I:C ratio is used to allow the model to better capture the observed iodide concentrations.

Results and Discussion

Overview – Global scale trends

281 In total, 127 sea surface iodide observations and 130 sea surface iodate observations were
282 made during the three cruises, including two time series. Measurements were made at 98
283 different sampling locations, spanning latitudes from ~12°N to ~70°S. This is a substantial
284 increase in data coverage for the Indian Ocean and the Indian sector of the Southern Ocean
285 region, which was previously particularly lacking in observations of sea surface iodine
286 speciation (Chance et al. 2014). As noted in the introduction, only two sea surface iodide
287 observations for the Indian Ocean were included in Chance et al., 2014, and both were from
288 the Arabian Sea (see Figure 1).
289
290   *[Figure 2]*
291
292   *[Figure 3]*
293
294 The lowest iodide concentrations were observed at high latitudes, while the highest
295 concentrations were encountered at the northern extent of the southern sub-tropical region
296 and within the tropics (Figure 2). This latitudinal trend in sea surface iodide concentration
297 broadly follows those observed in other ocean basins (Figure 3). In addition, a 'dip' in sea
298 surface iodide concentrations is seen around the equator and elevated concentrations are seen
299 in coastal polar waters, as observed elsewhere (Figures 2 & 3) (Chance et al. 2014; Chance et
300 al. 2010). While these global scale trends are well documented in the Atlantic and Pacific
301 basins e.g.(Bluhm et al. 2011; Campos, Sanders, and Jickells 1999; Truesdale, Bale, and
302 Woodward 2000; Tsunogai and Henmi 1971), to the best of our knowledge this is the first
303 time they have been confirmed in the Indian Ocean and the Indian section of the Southern
304 Ocean.
305
306   *[Figure 4]*
307
308 Considering the entire data set, sea surface iodide concentrations ranged from 20 to 1241 nM,
309 with a median of 61 nM (Figure 4). The very large range in the data is primarily due to the
310 presence of two very high outliers in the Bay of Bengal, which are discussed later. When
311 these outliers are excluded, the upper limit of the data is reduced to 250 nM, bringing the
312 range within the global range of marine iodide concentrations previously reported (Chance et
313 al. 2014). The large range in the overall data set can mainly be ascribed to the large span of
314 latitudes covered. The median value is somewhat lower than the global median value (77 nM;
315 (Chance et al. 2014), reflecting the bias towards high latitude/low iodide samples in our data
316 set.
317
318   *[Figure 5]*
319
320 Sea surface iodate concentrations ranged from 51 to 495 nM, with a median of 294 nM.
321 Iodate concentrations broadly showed the opposite pattern to iodide concentrations, with
322 highest median values in the Southern Ocean (median of 323 nM), intermediate values in the
323 subtropical Indian Ocean (median of 294 nM) and lowest values in the tropical Indian Ocean
324 (median 196 nM). However, there was only a very weak, inverse linear relationship between
325 sea surface iodide and iodate concentrations ($R^2 = 0.16$, $p = 3 \times 10^{-6}$; Figure 5). Total iodine
326 concentrations in seawater are typically ~450-500 nM (Chance et al. 2014, and references
327 therein), with the budget dominated by iodide and iodate. Here we find the sum of iodide and
328 iodate was less than this at most sampling locations (Figure 5), with a median value of 380
329 nM (range 88 to 560 nM). Although somewhat unusual, comparable low total inorganic
330 iodine concentrations have been reported elsewhere (e.g. North Sea (Hou et al. 2007), South

East China Sea (Wong and Zhang 2003)). Depleted total inorganic iodine may be due to uptake of iodine to the particulate phase in the surface ocean, or the presence of a significant dissolved organic iodine reservoir. Although it could imply substantial loss of iodine from the surface ocean to the atmosphere, current knowledge suggests the magnitude of such fluxes (e.g. Carpenter et al. 2013) is too small to have such a large impact on the sea surface concentrations. No clear relationships were evident between total inorganic iodine and either latitude or nitrate concentration.

Our sampling area spanned a wide range of different hydrographic provinces and biogeochemical conditions (Figure 2). North of the equator, SST becomes a poor predictor of iodide concentration. To explore our data set further, we consider iodine speciation separately in each of three different hydrographic regimes (tropical, mid-latitudes and polar).

South Indian subtropical gyre (~23-42°S)

In the southern Indian Ocean, samples were collected along an approximate latitudinal transect at 57°E, from ~23-42°S. The open ocean transect spanned the Indian South subtropical gyre and the subtropical convergence zone. North of this region, at ~15°S, the Southern Equatorial Current (SEC; not sampled in this work) has been noted as a clear biogeochemical front, separating subtropical gyre waters from the lower oxygen northern Indian Ocean (Grand et al. 2015). The subtropical gyre waters are characterised by very low nitrate concentrations and relatively high salinity (Figure 2), with SST rising from ~15°C in the south to ~29°C in the north. Between these latitudes, iodide concentrations ranged from 35 to 235 nM and increased in step with decreasing latitude and increasing SST (Figure 2). In this region, iodide is strongly correlated with both latitude ($R^2 = 0.86$, $p = 3 \times 10^{-20}$) and SST ($R^2_{adj} = 0.89$, $p = 2 \times 10^{-22}$) (Table 1). These are stronger correlations than the global relationships reported in Chance et al., 2014, and also have steeper gradients than found in the Chance et al. (2014) data set (Table 1).

Nitrate concentrations have been used to predict sea-surface iodide concentrations (Ganzeveld et al., 2009), and significant relationships between iodide and nitrate in upper 100 m of the water column have been reported for the subtropical Atlantic (Campos, Sanders, and Jickells 1999). However, in our data set no relationship between nitrate and iodide was apparent ($R^2 = 0.0004$; Table 1), implying sea surface nitrate concentrations are a poor predictor of sea surface iodide in this hydrographic region. This likely reflects the fact we only consider surface concentrations, which exhibited low nitrate concentrations (86% of data points were below 0.5 μM), but substantial gradients for iodide.

[Table 1]

Comparison of latitudinal sea surface iodide gradients between subtropical regions of different ocean basins

Using the extended dataset of global sea surface iodide observations (Chance et al., 2019), sea surface iodide gradients in the subtropical regions of each of the ocean basins were compared (Table 2). Considering each of the major ocean gyres individually reveals that the latitudinal dependency of iodide is greater in the Indian Ocean than in the north Atlantic, north Pacific or south Atlantic (Figure 3; Table 2). Iodide in the south Pacific appears to also have a steep gradient if observations from the Peruvian upwelling region (Cutter et al., 2018) are included in the analysis. However, this dataset has an unusually large iodide concentration range (153 to 790 nM) within a small latitudinal band (~11 to 16°S), and does

381 not exhibit a latitudinal trend in iodide as observed elsewhere. This distinct behaviour may be
382 due to the influence of sub-surface low oxygen waters. The remaining data from the south
383 Pacific subtropical gyre is from a transect along the 170°W meridian from 12 to 38°S
384 (Tsunogai and Henmi, 1971). If the Peruvian upwelling data is excluded from the gradient
385 analysis, the remaining data for the south Pacific exhibits a comparable iodide gradient to the
386 other ocean basins. The higher sea surface iodide gradient observed in the subtropical Indian
387 Ocean is driven by concentrations at low latitudes (~25°S) being approximately double those
388 at comparable latitudes in the other ocean basins, rather than a difference between
389 concentrations at the high latitude limits of the gyres (Figure 3).
390
391 The same trend is also evident in iodide vs. SST gradients, which are steeper in the Indian
392 Ocean than in other basins (Table 2). This implies that the differences between basins cannot
393 entirely be accounted for by differences in SST gradients. Examination of climatological data
394 (World Ocean Atlas, see Chance et al., 2014 for details) for the locations and months of
395 iodide observations does not demonstrate any clear differences in SST, nitrate or mixed layer
396 depth (MLD; defined using potential temperature) between ocean basins that might account
397 for the difference in iodide gradient. Although SST is not thought to directly impact iodine
398 cycling itself, its correlation with iodide concentration is important as it is widely used to
399 predict sea surface iodide fields for use in atmospheric models (e.g. Sarwar et al. 2016). The
400 stronger latitudinal dependency of iodide in the Indian Ocean that we observe here is not
401 replicated in sea surface iodide values predicted using commonly employed global scale
402 parametrisations based on sea surface temperature (Chance et al. 2014; MacDonald et al.
403 2014). This is likely to introduce uncertainties specific to the Indian Ocean when using such
404 parameterisations, for example in model calculations of iodine emissions from the sea
405 surface.
406
407   *[Table 2]*
408
409 The observational data is too limited to properly evaluate whether the pattern is the result of
410 seasonal biases in sampling; of the data used in this comparison, all subtropical Indian Ocean
411 observations were made during the southern hemisphere summer (January and February),
412 while the modal months for observations in other basins were November (North Atlantic),
413 October (South Atlantic) and March (North Pacific), see Table 2. In the South Pacific, if data
414 from the Peruvian upwelling is excluded, at least 90% of observations used were made in
415 January (Tsunogai and Henmi, 1971), i.e. in the same season as the Indian Ocean data.
416 Despite this, they have a lower latitudinal gradient (slope = 2.8 compared to 9.5; Table 2),
417 hinting that the difference between basins is not due to seasonal variation and/or sampling
418 biases and may instead be due to differences in iodine cycling.
419
420
421 Although less pronounced, a relatively steep iodide vs. latitude gradient in the Indian Ocean
422 is replicated in our model (Table 2), and hence a series of model sensitivity tests have been
423 conducted to investigate which processes may be responsible for the apparent differences in
424 the iodide gradient between ocean basins. Specifically, the model was run with each of the
425 iodide forcing processes in the Indian Ocean and Indian sector of the Southern Ocean
426 replaced by those from the Atlantic. Figure 6 shows the forcing fields used for the two ocean
427 basins, and the modelled iodide concentrations generated using these fields. The iodide vs.
428 latitude gradient is significantly increased between 30°S and 20°S when the Indian Ocean
429 productivity fields are replaced with those for the same latitudes in the Atlantic (Figure 6c).
430 This is driven by a smaller decrease in productivity with decreasing latitude in the Indian

431 Ocean than the Atlantic (Figure 6b). Thus productivity differences would act to decrease,
432 rather than increase the iodide gradient. In the model oxidation of iodide in the mixed layer
433 occurs as a function of nitrification, which is parameterised using the proportion of nitrate
434 regenerated in the mixed layer (see Wadley et al., 2020). Differences in this ratio between the
435 subtropical regions of the Indian and Atlantic oceans are small (Figure 6d), and have
436 negligible impact on modelled iodide in the Indian Ocean (Figure 6e). At lower latitudes
437 MLDs are similar in the two basins, but between ~35 and 45∘S, MLDs are significantly
438 deeper in the Atlantic due to the greater northward extent of polar waters (Figure 6f). Since a
439 deeper MLD dilutes iodide, modelled iodide concentrations are decreased between 45∘S and
440 35∘S when Atlantic MLDs are imposed in the Indian basin (Figure 6g). Again this further
441 increases the latitudinal gradient of iodide, rather than decreasing it. Precipitation and
442 evaporation act to dilute/concentrate iodide in the mixed layer, so changes in the net
443 freshwater flux with latitude could influence the latitudinal iodide gradient. However,
444 imposing the Atlantic surface freshwater flux in the Indian Basin had a negligible effect on
445 modelled iodide (Figure 6i). Finally, the I:C ratio in the model determines the amount of
446 iodide produced per unit carbon fixed by primary production. To fit model to the observed
447 iodide in each basin, a higher I:C ratio is utilised in the Indian than in the Atlantic Ocean
448 (Figure 6j; see Wadley et al., 2020). Replacing the I:C ratio in the Indian Ocean with that for
449 the Atlantic Ocean results in lower modelled iodide concentrations in the Indian Ocean, but
450 little overall change in the latitudinal iodide gradient (Figure 6k). Note the model currently
451 underestimates the iodide gradient in the Indian Ocean (Table 2), and further refinement of
452 the spatial variation in the I:C ratio is needed to improve the model fit to observations. This
453 may show that the I:C ratio influences latitudinal gradients of iodide.
454
455 In summary, basin scale differences in four key forcing fields (productivity, nitrification,
456 MLD and P-E) do not appear to explain the difference in latitudinal iodide gradient between
457 the subtropical Indian and southern Atlantic oceans. In fact, differences in productivity and
458 MLD between the two basins appear to have the opposite effect, as Atlantic forcing fields
459 resulting in an even steeper gradient than Indian Ocean forcing fields. Removal of advection
460 in the model also results in an increase rather than a decrease in iodide at sub-tropical
461 latitudes in the Indian Ocean (see later/Figure 7) and hence an even steeper latitudinal iodide
462 gradient. This suggests the basin scale differences in iodide gradient are not due to advection
463 in the Indian Ocean. Instead we suggest the difference may be the result of different
464 phytoplankton types, with differing I:C iodide production ratios between basins. The need for
465 a higher I:C ratio in the Indian Ocean/Indian sector of the Southern Ocean than the Atlantic
466 (Figure 6j) implies there may be profound differences in microbiological community
467 composition, and/or the cycling of iodine by such organisms between basins. *Synechococcus*
468 dominates in the subtropical Indian Ocean, whereas Nanoeucaryotes and *Prochlorococcus*
469 dominate in the subtropical Atlantic (Alvain et al., 2008). Recent work by our group indicates
470 that the I:C ratio associated with iodide production varies between phytoplankton types, with
471 *Synechococcus* having a higher I:C ratio than Nanoeucaryotes and *Prochlorococcus* types
472 (Wadley et al., *Manuscript in Preparation*). Hence the higher relative abundance of
473 *Synechococcus* in the subtropical Indian Ocean might account for the higher iodide
474 concentrations observed at low latitudes, and the resulting steeper sea-surface iodide gradient.
475 This also indicates that any change in microbiological community composition associated
476 with climate change could significantly impact ocean iodide concentrations.
477
478   *[Figure 6]*
479
480

Southern Ocean domain (~42˚S - 68˚S)

At the subtropical front around 42˚S, a decrease in salinity is accompanied by a sharp increase in nitrate concentration (Figure 2). This front marks the transition from the Indian Ocean to the Southern Ocean (Orsi, Whitworth, and Nowlin 1995). The colder, nutrient rich waters south of the subtropical front have much lower iodide content, with a median concentration of 43 nM and range of 20 to 104 nM (Figure 4). Relationships between iodide concentrations and latitude or SST in this region are much weaker than those observed further north (Table 1). This may be due to disruption by strong, but variable, vertical mixing events, which are characteristic of the Southern Ocean. No significant relationship between sea surface iodide and nitrate was observed in the Southern Ocean samples (Table 1).

The Southern Ocean samples may be further subdivided into coastal (i.e. near the Antarctic continent) and open ocean samples. The former category was defined as samples falling within the Austral Polar biogeochemical province (Longhurst 1998), while the latter category included samples from both the Antarctic and Sub-Antarctic provinces, and spanned the Polar Front. The range and distribution of concentrations seen in the coastal and open ocean subsets are very similar (Figure 4), despite the coastal samples spanning a much narrower latitudinal range (69 - 64∘ S, compared to 63 - 43∘ S). In the open ocean sub-set, iodide concentrations generally decrease moving south (R^2 = 0.16; p = 0.04), while in the coastal samples at the most southerly extreme of the data set, this relationship breaks down (R^2 = 0.02; Figure 2). A similar pattern is also reported in the global compilation of Chance et al. (2014), where Southern Ocean and Antarctic samples were predominantly from the Atlantic sector and the western Antarctic Peninsula. The range of concentrations observed in the Antarctic coastal samples (20 to 95 nM) is within that observed previously in coastal Antarctic waters during the austral summer (Chance et al. 2010). The magnitude of this variability is greater than can be accounted for by normalizing the iodide concentrations to salinity variations (e.g. due to ice melt water).

Northern Indian Ocean including southern Bay of Bengal (~4∘S - 11∘N)

Samples were collected from open ocean locations in the Bay of Bengal and the Arabian Sea, between ~4∘S and ~11∘N. Sampling locations were all within the Indian Monsoon Gyre biogeochemical province (Longhurst 1998), and shared high sea-surface temperatures, variable salinity and typically low nitrate concentrations. This sample sub-set showed the highest, and most variable iodide concentrations (range 63 to 1241 nM, median 165 nM; Figure 4). This was primarily due to the occurrence of two extremely high iodide concentrations (>900 nM) in the Bay of Bengal; excluding these values, the range in tropical open ocean iodide was still the largest of the three hydrographic regions surveyed, with a maximum of 250 nM being observed in the Bay of Bengal (Figure 4). Such concentrations are comparable to measurements made at similar latitudes elsewhere (Figure 3).

All but one of the samples collected around the equator (~5∘S to ~3∘N) during the SK333 cruise had iodide concentrations in the range ~80 to 120 nM. The exception to this was a station at 0∘N where iodide concentrations reached ~200 nM (Figure 2). As iodide concentrations were typically lower than in tropical waters further north and south (Figure 2), there was a positive rather than negative correlation between iodide and absolute latitude in this sample set (Table 1). A similar dip at very low latitudes is also seen in the Atlantic and Pacific (Figure 3; Chance et al., 2014). This feature is replicated in our model and is thought to be due to the advection and vertical mixing associated with the seasonally varying equatorial current system (see final section).

531 As already noted, exceptionally high sea surface iodide concentrations of 1241 and 949 nM
532 were observed at two stations in the Bay of Bengal (BS3 & BS8). Very high near-surface
533 iodide concentrations were also observed at Station AR nearby (2039, 1546 & 479 nM at 10,
534 25 & 50 m respectively; see Figure S1 in Supplementary Information). We believe these
535 concentrations are real, and not the result of contamination or analytical error because: (i)
536 repeat analyses gave the same results, (ii) very high concentration samples were also
537 analysed by ion chromatography, a completely independent method, and this yielded
538 concentrations within 10% of those obtained by voltammetry (1277 and 960 nM for samples
539 BS3 and BS8 respectively, and 1693 nM for Station AR at 25 m depth), (iii) results for
540 Station AR show oceanographic consistency (see Figure S1), (iv) no likely sources of iodine
541 contamination were present during sampling. It is possible that the high, very localised,
542 iodide concentrations could arise from the break-down of an iodine rich substrate, for
543 instance a mass of brown macroalgae such as *Laminaria digitata*. However, nothing unusual
544 of this nature was observed during sampling. These very high iodide levels result in total
545 (dissolved) inorganic iodine concentrations more than double (2.2 and 2.6 times larger) the
546 near universally observed value of ~450-500 nM (Chance et al. 2014). This amount of iodide
547 cannot be accounted for solely by the reduction of iodate in the water column (as this could
548 only yield concentrations up to ~500 nM). Instead it implies an exogenous source of iodine,
549 which is either already in the form of iodide, or is reduced to iodide following introduction to
550 the water column.
551
552 Samples with elevated iodide exhibited relatively low surface salinity (Figure S1,
553 Supplementary Information), possibly indicating an association with freshwater inputs. The
554 Bay of Bengal is characterized by very heterogenous salinity - low salinity is caused by
555 monsoon rainfall and high riverine inputs to the north, while high salinity water arrives from
556 the Arabian Sea via the intense Southwest Monsoon Current (SMC). The BoBBLe cruise
557 took place during the Asian summer monsoon season, during which the region experiences
558 high rainfall. However, atmospheric wet deposition is thought unlikely to be the source of
559 elevated iodide because meteoric water has a lower iodine content than seawater (20-124 nM
560 (Sadasivan and Anand 1979); 4.7-26.2 nM; (Gilfedder, Petri, and Biester 2007)).
561 Furthermore, observations of elevated sea surface iodide did not correspond to rainfall events
562 encountered during the BoBBLe cruise itself. Similarly, riverine inputs are unlikely to be the
563 iodide source, as total iodine concentrations in rivers, including the Ganges, are lower than or
564 comparable to those in seawater ($\leq$ 20 ug/L, ~ 157 nM; (Moran, Oktay, and Santschi 2002;
565 Ghose, Das, and Saha 2003)), and the area surveyed was away from major outflows. While
566 freshwater inputs are thus not thought to be the source of the excess iodide, stratification
567 caused by rainwater dilution of surface layers could plausibly contribute to the persistence of
568 high iodide concentrations at the ocean surface.
569
570 In addition to the SMC, the second main oceanographic feature in the BoBBLe study area is a
571 wind driven upwelling feature called the Sri Lanka Dome (SLD), which manifests as a large
572 cyclonic gyre in the south western part of the BoB to the east of Sri Lanka. Both the SLD and
573 the SMC were well developed and distinct during the study period (Vinayachandran et al.
574 2018). Extreme iodide values were observed at stations north (AR, BS3) and east (BS8) of
575 the SLD, but not in the stations that were inside the SLD (Figure S1 and S2, Supp Info)
576 (Vinayachandran et al. 2018). High iodide station AR exhibited high seawater pCO_2 (467–
577 554 atm), low surface pH and low alkalinity, indicative of upwelled waters that are presumed
578 to be associated with the SMC (Vinayachandran et al. 2018). Station BS3 was close in space
579 and time, while station BS8 was also influenced by the SMC (Vinayachandran et al. 2018).
580 We therefore speculate that the high iodide observed is in some way related to the SMC.

581   However, the high salinity core of the SMC (i.e. Arabian Sea water), which was evident at
582   depths of ~25-150 m at stations east of the SLD, was not itself associated with elevated
583   iodide (Figure S1, Supplementary Information). This suggests the excess iodide was not in
584   the main water mass carried by the SMC, and did not originate from the Arabian Sea. Instead
585   we suggest its source could be coastal waters that are entrained in the periphery of the SMC
586   and carried into the BoB with it.

588   Comparable high iodide concentrations have previously been observed in low oxygen sub-
589   surface waters in the north western part of the Arabian Sea (Farrenkopf and Luther 2002);
590   these were attributed to advection from shelf sediments. More recently, plumes of very high
591   iodide sub-surface concentrations (~1000 nM) have also been reported in sub-oxic waters in
592   the Eastern Tropical South Pacific (ETSP) (Cutter et al. 2018) and the Eastern Tropical North
593   Pacific (ETNP) (Moriyasu et al. 2020). In both cases some outcropping of elevated iodide
594   concentrations at the ocean surface was observed (Cutter et al., 2018; Moriyasu et al., 2020).
595   In the ETSP, the plume followed an isopycnal and was associated with corresponding
596   features in Fe(II) and nitrite, so was again thought to be due to a shelf sediment iodide source
597   (Cutter et al. 2018). Elevated concentrations persisted more than 1000 km from the shelf
598   break due to the relatively long iodide lifetime in seawater with respect to oxidation. Trace
599   metal concentrations were not measured during the BoBBLe cruise, but sub-surface waters in
600   the region have previously been reported to contain additional dissolved Fe from sedimentary
601   inputs (Grand et al. 2015), suggesting it is plausible that stations on the edge of the SLD
602   could be influenced by sedimentary interactions. Waters over the western Indian shelf
603   experience severe hypoxia (Naqvi et al. 2000), and so could be subject to significant
604   sedimentary iodide inputs as implicated in other low oxygen regions (Cutter et al. 2018;
605   Farrenkopf and Luther 2002). During the Southwest Monsoon, the West Indian Coastal
606   Current flows south along the west coast of India, and these waters are advected along the
607   path of the SMC into the BoB. We speculate that such waters have potential to contain
608   'excess' iodide levels as a result of sedimentary inputs, and that it is the remnants of these
609   water masses that we may have sampled. Iodide has a long lifetime in oxygenated seawater
610   (6 months to several years; Chance et al., 2014; Hardisty et al., 2020), and so a signal may be
611   expected to persist where other indicators of sedimentary inputs are lost (Cutter et al., 2018),
612   and could potentially reach the ocean surface via upwelling areas such as the south western
613   BoB (Figure S2, Supplementary Information).

615   In the northern Indian Ocean agreement between model and observations is poorer than
616   elsewhere, but modelled concentrations fall within the range of the observations in the area.
617   Furthermore, the model predicts unusually high iodide concentrations in the BoB, where
618   extremely high iodide concentrations were observed. As the model does not currently include
619   sedimentary processes, this suggests additional physical and biogeochemical processes may
620   also contribute to elevated iodide levels in the BoB.

622   We believe this is the third reported observation of very elevated (>500 nM) iodide
623   concentrations at the surface of the open ocean. The dataset reported by Cutter et al. (2018)
624   contains two surface samples with iodide concentrations of 594 and 960 nM immediately off
625   the coast of Peru, while transects in (Moriyasu et al. 2020), show patches of high iodide
626   outcropping at the surface. An extreme surface iodide concentration of ~700 nM has also
627   been reported for the brackish waters of the Skaggerrak (Truesdale, Danielssen, and Waite
628   2003), a shallow strait that is subject to hypoxia (Johannessen and Dahl 1996). The paucity of
629   previous observations suggests it is a rare phenomenon, but it may nonetheless be of local
630   significance.

631
632 Atmospheric ozone deposition and iodine emission fluxes will proportionally increase where
633 surface iodide concentrations are high. An increase in iodide from 150 to 2050 nM (x 13.7)
634 leads to a 5-6 fold increase in total iodine emissions, for a typical ambient ozone
635 concentration of 25 ppb and a wind speed of 7 m s^{-1} (Figure S3, Supplementary Information).
636 This comprises a 4.5-fold increase in HOI emissions, which dominates the flux, and a 30-fold
637 increase in I$_2$ emissions, which increase from 5.5% of the total flux at 150 nM iodide to 28%
638 at 2050 nM iodide. It is assumed that the regional atmospheric impact of such 'hot-spots' will
639 be low, as they will only represent a small proportion of the relevant footprint area. However,
640 the atmospheric impacts of such areas may become significant if either very localised
641 processes are being considered, or their extent and/or frequency of occurrence increases. As
642 areas of low oxygen waters in contact with shelf sediments become more extensive (Naqvi et
643 al. 2000), the possibility that this could impact on surface iodide concentrations in coastal
644 regions may need to be considered. Understanding the potential impact of low oxygen
645 conditions and sedimentary inputs on surface iodide concentrations, and hence local
646 atmospheric chemistry, requires both the sedimentary fluxes and the lifetime of iodide in
647 oxygenated seawater to be better constrained. The atmospheric boundary layer above the
648 northern Indian Ocean has high iodine oxide (IO) levels, with inputs dominated by the
649 inorganic iodine flux from the ozone-iodide reaction (>90%), as a result of "ozone-related"
650 pollution outflow (Prados-Roman et al. 2015). Hence atmospheric chemistry in the region
651 may be particularly sensitive to changes in the surface iodide budget.
652
653 ***Exploring physical and biogeochemical controls on sea surface iodide in the Indian Ocean***
654 ***using the ocean iodine cycling model***
655
656 The trends in sea surface iodide concentrations observed across the south Indian subtropical
657 gyre and Indian sector of the Southern Ocean are well replicated by the iodine cycling model
658 of Wadley et al. (2020) (Figure 7a). We used the model to explore how physical (MLD,
659 advection, net surface freshwater flux) and biogeochemical (productivity, iodide formation to
660 carbon fixation ratio) factors influence iodine cycling in the study area.
661
662 Biological productivity drives iodide formation in the model, but has a different spatial
663 distribution to iodide in the Indian Ocean basin (Figure 7a, b). Although both tend to
664 increase moving north along the transect, the very strong peak in productivity centred on 40 S
665 (Figure 7b) has no corresponding peak in either observed or simulated iodide concentrations.
666 This indicates that other processes are also important in determining the iodide distribution.
667 Imposing a constant level of productivity (set at the annual mean for each grid point)
668 throughout the year had very little effect on modelled iodide concentrations, (Figure 7c),
669 suggesting that the seasonal cycle of iodide production does not have a dominant effect on
670 iodide concentrations. Halving the productivity in the model reduces simulated iodide
671 concentrations by around a half south of 40 S (where iodide is very low, and iodate high), but
672 only by around a quarter north of 40 S, and has almost no effect in the northern Indian Ocean
673 where modelled iodide is highest (and thus modelled iodate lowest, at ≤ 300 nM). This may
674 indicate that lower iodate concentrations to the north limit iodide production. The amount of
675 iodide produced per unit primary productivity is specified in a spatially variable I:C ratio
676 (Figure 7e; (Wadley et al. 2020). Halving this ratio is equivalent to halving primary
677 production, and has identical consequences (Figure 7f).
678
679 As iodide is assumed to be evenly distributed throughout the mixed layer, surface
680 concentrations are dependent on the MLD. Summer MLD minima are similar throughout the

681 section, with typical values of a few tens of metres, whereas late winter MLD maxima
682 increase with latitude (Figure 7j). Deepening of the MLD decreases iodide concentrations
683 throughout the mixed layer as a result of dilution, while shoaling of the MLD decreases the
684 total amount of iodide present integrated over the mixed layer, but does not change the
685 concentration at any given depth within it. Removing the seasonal cycle of MLD in the
686 model (by replacing with the annual mean MLD for each grid point) increases surface iodide
687 concentrations, particularly at lower latitudes (Figure 7k), indicating that the removal of
688 iodide through MLD shoaling is important. Removing both seasonal and spatial variation in
689 the MLD by setting it to a uniform 56 m results in substantial increases in surface iodide
690 concentrations at higher latitudes (Figure 7l), due to elimination of both dilution and shoaling
691 effects. In contrast, doubling the MLD decreases iodide concentrations, with the greatest
692 effect seen where iodide concentrations are highest (Figure 7m). Interestingly, this brings
693 modelled and observed iodide concentrations into good agreement around the equator.
694 Inspection of SK333 CTD data indicates that at the time of sampling, actual MLDs in this
695 region (between 50 to 100 m) were deeper than the climatological values used in the model
696 (less than 40 m; Figure 7j), which may indicate that short term variations in MLD, for
697 example due to weather events, affected iodide concentrations.
698
699 In the model, phytoplankton mediated iodide formation occurs 60 days after the uptake of
700 carbon, and the interplay of this lag period with the MLD cycle could influence iodide
701 concentrations. However, changing the duration of the lag to 0 and 120 days in the model has
702 only a small impact on the iodide, and only at high latitudes (Figures 7g and 7h).
703
704 The seasonal MLD cycle imposes an annual timescale on the removal of iodide from the
705 mixed layer where the seasonal cycle has a large amplitude, but vertical and horizontal
706 advection also exchange water between the surface layer and ocean interior. Turning off the
707 circulation in the model, so that vertical mixing is the only physical mechanism for iodide
708 removal from the mixed layer also results in increased mixed layer iodide concentrations at
709 almost all locations (Figure 7n).
710
711 In addition to removal by MLD shoaling, the model also allows for the removal of mixed
712 layer iodide by oxidation to iodate, via a pathway linked to nitrification (Hughes et al. 2020;
713 Wadley et al. 2020). Eliminating this pathway has little impact on modelled iodide
714 concentrations at high latitudes, but results in a significant increase in concentrations north of
715 40◦S (Figure 7i). This is because the iodide oxidation timescale in the model is multi-annual,
716 and hence its impact is modulated by the lifetime of iodide in the mixed layer with reference
717 to vertical mixing. At high latitudes, large seasonal changes in the MLD result in low
718 sensitivity to oxidation, whereas at lower latitudes there is less annual mixed layer exchange,
719 resulting in a longer iodide residence time, and hence oxidation is a more important process
720 for mixed layer iodide removal.
721
722 The net surface freshwater flux resulting from precipitation and evaporation (P-E) acts to
723 respectively dilute or concentrate iodide in the mixed layer (Figure 7o). Setting the P-E flux
724 to zero has a small impact on iodide, decreasing concentrations in the subtropical gyres where
725 evaporation is strong. Thus at these latitudes evaporation results in a modest increase in
726 iodide concentrations.
727
728 The model sensitivity tests described above confirm that sea surface iodide concentrations in
729 the Indian and Southern Oceans are determined by a combination of factors which interact
730 non-linearly. The dominant processes that determine iodide concentrations are the rate of

731 iodide production, which is a function of productivity and the I:C ratio, and the MLD and its
732 seasonal cycle, which acts to dilute and remove iodide from the surface layer. Loss by
733 oxidation has the greatest impact at lower latitudes where physical removal mechanisms are
734 weakest. As the climate changes over coming decades, changes in any of these factors are
735 likely to result in changes in sea surface iodide concentrations. For example, the 100 to 300
736 m depth layer in the Indian Ocean has significantly warmed since 2003 as a result of heat
737 distribution from the Pacific (Nieves et al., 2015) and this is likely to reduce stratification,
738 and enhance vertical mixing, with an accompanying reduction in mixed layer iodide
739 concentrations. Conversely, changes in nutrient inputs to the Indian Ocean and declining
740 oxygen levels will act to reduce iodide oxidation, due to its association with nitrification
741 (Hughes et al. 2020), and hence may act to increase iodide concentrations. Any changes in
742 productivity, and/or shifts to different phytoplankton types with different I:C ratios, will also
743 potentially alter the rate of iodide production, although this has been found to predominantly
744 impact on iodide concentrations at higher latitudes (Wadley et al., *Manuscript In*
745 *Preparation*).
746
747
748 *[Figure 7]*
749
750
751
752 **Concluding remarks**
753 The 127 sea surface iodide observations examined here represent a substantial (>10%)
754 increase in the number of measurements that have been made across the global oceans (925
755 individual observations were included in Chance et al., 2014). They span nearly 80 degrees
756 latitude in the Indian Ocean and Indian sector of the Southern Ocean, a region where very
757 few (n=2) surface observations were previously available. This increase in observations will
758 facilitate better predictions of sea surface iodide concentrations, and consequent atmospheric
759 chemistry, and the data has already been incorporated in a new global iodide database
760 (Chance et al., 2019) and parameterisation (Sherwen et al. 2019).
761
762 The large scale latitudinal trends observed in the Indian Ocean and Indian sector of the
763 Southern Ocean are similar to those in other ocean basins (Chance et al. 2014). At subtropical
764 latitudes, the latitudinal and temperature dependency of iodide is steeper than in other ocean
765 basins. Therefore, using global scale relationships with SST (e.g. (MacDonald et al. 2014)) to
766 predict regional scale iodide concentrations for the Indian Ocean will be subject to biases.
767 Exploration of this basin scale difference using the a state-of-the-art global iodine cycling
768 model (Wadley et al. 2020) indicates that it may be driven by differences in biological iodide
769 production. We have also used the model to explore the controls on the sea-surface iodide
770 distribution in the Indian Ocean basin. Model sensitivity tests indicate that sea surface iodide
771 is likely to be a function of vertical mixing and the seasonal cycle in MLD, the rate of iodide
772 production, which is related to productivity and the I:C ratio, and – at lower latitudes – iodide
773 oxidation in the mixed layer. These factors interact in a non-linear manner.
774
775 Exceptionally high iodide concentrations were observed at a small number of stations in the
776 northern Indian Ocean. Such high concentrations are rare, but have been reported previously
777 in low oxygen subsurface waters (Farrenkopf and Luther 2002), and more recently, in surface
778 waters above the Peruvian and Mexican oxygen deficient zones (Cutter et al. 2018; Moriyasu
779 et al. 2020). In all cases, the excess iodide is suggested to be sedimentary in origin, raising
780 the possibility that processes at the sea floor could influence air-sea interactions, should that

781 water reach the sea surface. Although such iodide 'hot-spots' are unlikely to have significant
782 impact on a global or regional scale, it is possible they may impact on local scale air-sea
783 exchange processes involving iodine. Their existence also means care must be taken to
784 ensure iodide concentrations used to generate parameterisations are representative of the
785 entire study area.
786
787 Marine iodine cycling is anticipated to be affected by global change, with consequent impacts
788 on atmospheric chemistry. The Indian Ocean basin is subject to a number of specific
789 pressures with potential to affect iodine cycling. Despite the empirical relationship between
790 SSI and SST, changes in vertical mixing as a result of the Indian Ocean warming may in fact
791 reduce mixed layer iodide concentrations. Meanwhile, changes to biological processes as a
792 result of anthropogenic nutrient inputs, ocean deoxygenation and changes in heat distribution
793 are likely to impact iodide production and loss processes. Such changes will in turn impact
794 ozone deposition and iodine emission from the sea surface.

References

Aksenov, Yevgeny, Sheldon Bacon, Andrew C. Coward, and A. J. George Nurser. 2010. 'The North Atlantic inflow to the Arctic Ocean: High-resolution model study', Journal of Marine Systems, 79: 1-22.

Allan, J. D., P. I. Williams, J. Najera, J. D. Whitehead, M. J. Flynn, J. W. Taylor, D. Liu, E. Darbyshire, L. J. Carpenter, R. Chance, S. J. Andrews, S. C. Hackenberg, and G. McFiggans. 2015. 'Iodine observed in new particle formation events in the Arctic atmosphere during ACCACIA', Atmospheric Chemistry and Physics (Print), 15: 5599-609.

Behrenfeld, Michael, and Paul Falkowski. 1997. 'Behrenfeld MJ, Falkowski PG. Photosynthetic rates derived from satellite-based chlorophyll concentration. Limnol Oceanogr 41: 1-20', Limnology and Oceanography, 42: 1-20.

Bluhm, K., P. L. Croot, O. Huhn, G. Rohardt, and K. Lochte. 2011. 'Distribution of iodide and iodate in the Atlantic sector of the southern ocean during austral summer', Deep-Sea Research Part II-Topical Studies in Oceanography, 58: 2733-48.

Bluhm, K., P. Croot, K. Wuttig, and K. Lochte. 2010. 'Transformation of iodate to iodide in marine phytoplankton driven by cell senescence', Aquatic Biology, 11: 1-15.

Campos, M.L.A.M. 1997. 'New approach to evaluating dissolved iodine speciation in natural waters using cathodic stripping voltammetry and a storage study for preserving iodine species', Marine Chemistry, 57: 107-17.

Campos, M.L.A.M., A. M. Farrenkopf, T. D. Jickells, and G. W. Luther. 1996. 'A comparison of dissolved iodine cycling at the Bermuda Atlantic Time-Series station and Hawaii Ocean Time-Series Station', Deep-Sea Research Part II: Topical Studies in Oceanography, 43: 455-66.

Campos, M.L.A.M., R. Sanders, and T. Jickells. 1999. 'The dissolved iodate and iodide distribution in the South Atlantic from the Weddell Sea to Brazil', Marine Chemistry, 65: 167-75.

Carpenter, L. J., S. M. MacDonald, M. D. Shaw, R. Kumar, R. W. Saunders, R. Parthipan, J. Wilson, and J. M. C. Plane. 2013. 'Atmospheric iodine levels influenced by sea surface emissions of inorganic iodine', Nature Geoscience, 6: 108-11.

Chance, R., A. R. Baker, L. Carpenter, and T. D. Jickells. 2014. 'The distribution of iodide at the sea surface', Environmental Science-Processes & Impacts, 16: 1841-59.

Chance, R.J., L. Tinel, T. Sherwen, A. Baker, T. Bell, J. Brindle, M.L.A.M. Campos, P. Croot, H. Ducklow, P. He, B. Hoogakker, F.E. Hopkins, C. Hughes, T. Jickells, D. Loades, D.A. Reyes Macaya, A.S. Mahajan, G. Malin, D.P. Phillips, A.K. Sinha, A. Sarkar, I.J. Roberts, R. Roy, X. Song, H.A. Winklebauer, K. Wuttig, M. Yang, P. Zhou, and L.J. Carpenter. 2019. "Global sea-surface iodide observations, 1967-2018." In. British Oceanographic Data Centre - Natural Environment Research Council, UK.

Chance, R.J., L. Tinel, T. Sherwen, A.R. Baker, T. Bell, J. Brindle, M.L.A.M. Campos, P. Croot, H. Ducklow, P. He, F. Hopkins, B. Hoogakker, C. Hughes, T.D. Jickells, D. Loades, D.A.R. Macaya, A.S. Mahajan, G. Malin, D. Phillips, I. Roberts, R. Roy, A. Sarkar, A.K. Sinha, X. Song, H. Winkelbauer, K. Wuttig, M. Yang, Peng Zhou, and L.J. Carpenter. 2019. 'Global sea-surface iodide observations, 1967-2018', Scientific Data, 6: 286.

Chance, Rosie, Keith Weston, Alex R. Baker, Claire Hughes, Gill Malin, Lucy Carpenter, Michael P. Meredith, Andrew Clarke, Timothy D. Jickells, Paul Mann, and Helen Rossetti. 2010. 'Seasonal and interannual variation of dissolved iodine speciation at a coastal Antarctic site', Marine Chemistry, 118: 171-81.

875 Cuevas, C. A., N. Maffezzoli, J. P. Corella, A. Spolaor, P. Vallelonga, H. A. Kjaer, M.
876   Simonsen, M. Winstrup, B. Vinther, C. Horvat, R. P. Fernandez, D. Kinnison, J. F.
877   Lamarque, C. Barbante, and A. Saiz-Lopez. 2018. 'Rapid increase in atmospheric
878   iodine levels in the North Atlantic since the mid-20th century', Nature
879   Communications, 9: 6.
880 Cutter, G. A., J. G. Moffett, M. C. Nielsdottir, and V. Sanial. 2018. 'Multiple oxidation state
881   trace elements in suboxic waters off Peru: In situ redox processes and
882   advective/diffusive horizontal transport', Marine Chemistry, 201: 77-89.
883 Ducklow, H. W., M. R. Stukel, R. Eveleth, S. C. Doney, T. Jickells, O. Schofield, A. R. Baker,
884   J. Brindle, R. Chance, and N. Cassar. 2018. 'Spring-summer net community production,
885   new production, particle export and related water column biogeochemical processes in
886   the marginal sea ice zone of the Western Antarctic Peninsula 2012-2014', Philosophical
887   Transactions of the Royal Society a-Mathematical Physical and Engineering Sciences,
888   376: 15.
889 Edwards, A., and V. W. Truesdale. 1997. 'Regeneration of inorganic iodine species in Loch
890   Etive, a natural leaky incubator', Estuarine Coastal and Shelf Science, 45: 357-66.
891 Elderfield, H., and V. W. Truesdale. 1980. 'On the biophilic nature of iodine in seawater', Earth
892   and Planetary Science Letters, 50: 105-14.
893 Farrenkopf, A. M., and G. W. Luther. 2002. 'Iodine chemistry reflects productivity and
894   denitrification in the Arabian Sea: evidence for flux of dissolved species from
895   sediments of western India into the OMZ', Deep-Sea Research Part II-Topical Studies
896   in Oceanography, 49: 2303-18.
897 Farrenkopf, A. M., G. W. Luther, V. W. Truesdale, and C. H. Van Der Weijden. 1997. 'Sub-
898   surface iodide maxima: evidence for biologically catalyzed redox cycling in Arabian
899   Sea OMZ during the SW intermonsoon', Deep Sea Research Part II: Topical Studies in
900   Oceanography, 44: 1391-409.
901 Ganzeveld, L., D. Helmig, C. W. Fairall, J. Hare, and A. Pozzer. 2009. 'Atmosphere-ocean
902   ozone exchange: A global modeling study of biogeochemical, atmospheric, and
903   waterside turbulence dependencies', Global Biogeochemical Cycles, 23.
904 Ghose, N. C., K. Das, and D. Saha. 2003. 'Distribution of iodine in soil-water system in the
905   Gandak basin, Bihar', Journal of the Geological Society of India, 62: 91-98.
906 Gilfedder, B. S., M. Petri, and H. Biester. 2007. 'Iodine speciation in rain and snow:
907   Implications for the atmospheric iodine sink', Journal of Geophysical Research-
908   Atmospheres, 112: 7.
909 Grand, Maxime M., Christopher I. Measures, Mariko Hatta, William T. Hiscock, William M.
910   Landing, Peter L. Morton, Clifton S. Buck, Pamela M. Barrett, and Joseph A. Resing.
911   2015. 'Dissolved Fe and Al in the upper 1000 m of the eastern Indian Ocean: A high-
912   resolution transect along 95°E from the Antarctic margin to the Bay of Bengal', Global
913   Biogeochemical Cycles, 29: 375-96.
914 Hardacre, C., O. Wild, and L. Emberson. 2015. 'An evaluation of ozone dry deposition in global
915   scale chemistry climate models', Atmospheric Chemistry and Physics (Print), 15: 6419-
916   36.
917 Hardisty, D. S., T. J. Horner, S. D. Wankel, J. Blusztajn, and S. G. Nielsen. 2020. 'Experimental
918   observations of marine iodide oxidation using a novel sparge-interface MC-ICP-MS
919   technique', Chemical Geology, 532: 119360.
920 Hou, Xiaolin, Ala Aldahan, Sven P. Nielsen, Goran Possnert, Hartmut Nies, and Jim Hedfors.
921   2007. 'Speciation of I-129 and I-127 in seawater and implications for sources and
922   transport pathways in the North Sea', Environmental Science & Technology, 41: 5993-
923   99.

924 Hughes, C., E. Barton, H. Hepach, R. Chance, M. Pickering, and K. Hogg. 2020. 'Iodate
925   production in marine ammonium-oxidising bacteria', *Manuscript submitted to Global*
926   *Biogeochemical Cycles, manuscript number 2020GB006673.*
927 Jickells, T. D., S. S. Boyd, and A. H. Knap. 1988. 'Iodine cycling in the Sargasso Sea and the
928   Bermuda Inshore waters', Marine Chemistry, 24: 61-82.
929 Johannessen, T., and E. Dahl. 1996. 'Declines in oxygen concentrations along the Norwegian
930   Skagerrak coast, 1927-1993: A signal of ecosystem changes due to eutrophication?',
931   Limnology and Oceanography, 41: 766-78.
932 Longhurst, A.R. 1998. Ecological geography of the sea (Academic: San Diego).
933 Lu, Wanyi, Andy Ridgwell, Ellen Thomas, Dalton S. Hardisty, Genming Luo, Thomas J.
934   Algeo, Matthew R. Saltzman, Benjamin C. Gill, Yanan Shen, Hong-Fei Ling, Cole T.
935   Edwards, Michael T. Whalen, Xiaoli Zhou, Kristina M. Gutchess, Li Jin, Rosalind E.
936   M. Rickaby, Hugh C. Jenkyns, Timothy W. Lyons, Timothy M. Lenton, Lee R. Kump,
937   and Zunli Lu. 2018. 'Late inception of a resiliently oxygenated upper ocean', Science:
938   eaar5372.
939 Luther, G. W., C. B. Swartz, and W. J. Ullman. 1988. 'Direct determination of iodide in
940   seawater by cathodic stripping square-wave voltammetry', Analytical Chemistry, 60:
941   1721-24.
942 MacDonald, S. M., J. C. Gómez Martín, R. Chance, S. Warriner, A. Saiz-Lopez, L. J.
943   Carpenter, and J. M. C Plane. 2014. 'A laboratory characterisation of inorganic iodine
944   emissions from the sea surface: dependence on oceanic variables and parameterisation
945   for global modelling', Atmospheric Chemistry and Physics, 14: 5841-52.
946 Mahajan, Anoop S., Liselotte Tinel, Shrivardhan Hulswar, Carlos A. Cuevas, Shanshan Wang,
947   Sachin Ghude, Ravidas K. Naik, Rajani K. Mishra, P. Sabu, Amit Sarkar, N.
948   Anilkumar, and Alfonso Saiz Lopez. 2019. 'Observations of iodine oxide in the Indian
949   Ocean marine boundary layer: A transect from the tropics to the high latitudes',
950   Atmospheric Environment: X, 1: 100016.
951 McFiggans, G., H. Coe, R. Burgess, J. Allan, M. Cubison, M. R. Alfarra, R. Saunders, A. Saiz-
952   Lopez, J. M. C. Plane, D. J. Wevill, L. J. Carpenter, A. R. Rickard, and P. S. Monks.
953   2004. 'Direct evidence for coastal iodine particles from Laminaria macroalgae - linkage
954   to emissions of molecular iodine', Atmospheric Chemistry and Physics, 4: 701-13.
955 Moran, Jean E., Sarah D. Oktay, and Peter H. Santschi. 2002. 'Sources of iodine and iodine
956   129 in rivers', Water Resources Research, 38: 24-1-24-10.
957 Moriyasu, Rintaro, Zachary C. Evans, Kenneth M. Bolster, Dalton S. Hardisty, and James W.
958   Moffett. 2020. 'The Distribution and Redox Speciation of Iodine in the Eastern Tropical
959   North Pacific Ocean', Global Biogeochemical Cycles, 34: e2019GB006302.
960 Nakayama, E., T. Kimoto, K. Isshiki, Y. Sohrin, and S. Okazaki. 1989. 'Determination and
961   distribution of iodide-iodine and total-iodine in the north Pacific ocean by using a new
962   automated electrochemical method', Marine Chemistry, 27: 105-16.
963 Naqvi, S. W. A., D. A. Jayakumar, P. V. Narvekar, H. Naik, V. V. S. S. Sarma, W. D'Souza,
964   S. Joseph, and M. D. George. 2000. 'Increased marine production of N2O due to
965   intensifying anoxia on the Indian continental shelf', Nature, 408: 346.
966 Orsi, Alejandro H., Thomas Whitworth, and Worth D. Nowlin. 1995. 'On the meridional extent
967   and fronts of the Antarctic Circumpolar Current', Deep Sea Research Part I:
968   Oceanographic Research Papers, 42: 641-73.
969 Prados-Roman, C., C. A. Cuevas, T. Hay, R. P. Fernandez, A. S. Mahajan, S. J. Royer, M.
970   Galí, R. Simó, J. Dachs, K. Großmann, D. E. Kinnison, J. F. Lamarque, and A. Saiz-
971   Lopez. 2015. 'Iodine oxide in the global marine boundary layer', Atmospheric
972   Chemistry and Physics (Print), 15: 583-93.

973　Price, N.B. and Calvert, S.E., 1977. Contrasting geochemical behaviors of iodine and bromine
974　　　in recent sediments from the Namibian shelf. Geochimica et Cosmochimica Acta,
975　　　41(12): 1769-1775.
976　Rue, E.L., G.J. Smith, G.A. Cutter and K.W. Bruland, 1997, The response of trace element
977　　　redox couples to suboxic conditions in the water column, Deep Sea Res., Part I, 44,
978　　　113–134.
979　Sadasivan, S., and S. J. S. Anand. 1979. 'Chlorine, bromine and iodine in monsoon rains in
980　　　India', Tellus, 31: 290-94.
981　Sarwar, G., D. W. Kang, K. Foley, D. Schwede, B. Gantt, and R. Mathur. 2016. 'Technical
982　　　note: Examining ozone deposition over seawater', Atmospheric Environment, 141: 255-
983　　　62.
984　Schlitzer, R. 2014. "Ocean Data View." In.: http://odv.awi.de.
985　Schwehr, K. A., and P. H. Santschi. 2003. 'Sensitive determination of iodine species, including
986　　　organo-iodine, for freshwater and seawater samples using high performance liquid
987　　　chromatography and spectrophotometric detection', Analytica Chimica Acta, 482: 59-
988　　　71.
989　Sherwen, T., M. J. Evans, R. Sommariva, L. D. J. Hollis, S. M. Ball, P. S. Monks, C. Reed, L.
990　　　J. Carpenter, J. D. Lee, G. Forster, B. Bandy, C. E. Reeves, and W. J. Bloss. 2017.
991　　　'Effects of halogens on European air-quality', Faraday Discussions, 200: 75-100.
992　Sherwen, T.S., L. Tinel, R. Chance, L.J. Carpenter, and M.J. Evans. 2019. 'Re-approaching
993　　　global iodine emissions: A novel parameterisation for sea-surface iodide concentrations
994　　　using a machine learning approach. In prep.'.
995　Truesdale, V. W. 1978. 'Iodine in inshore and off-shore marine waters', Marine Chemistry, 6:
996　　　1-13.
997　Truesdale, V. W., A. J. Bale, and E. M. S. Woodward. 2000. 'The meridional distribution of
998　　　dissolved iodine in near-surface waters of the Atlantic Ocean', Progress in
999　　　Oceanography, 45: 387-400.
1000　Truesdale, V. W., D. S. Danielssen, and T. J. Waite. 2003. 'Summer and winter distributions
1001　　　of dissolved iodine in the Skagerrak', Estuarine Coastal and Shelf Science, 57: 701-13.
1002　Truesdale, V. W., and C. P. Spencer. 1974. 'Studies on the determination of inorganic iodine
1003　　　in seawater', Marine Chemistry, 2: 33-47.
1004　Tsunogai, S. 1971. 'Iodine in the deep water of the ocean', Deep-Sea Research, 18: 913-19.
1005　Tsunogai, S., and T. Henmi. 1971. 'Iodine in the surface water of the Pacific Ocean', Journal
1006　　　of the Oceanographical Society of Japan, 27: 67-72.
1007　Vinayachandran, P. N., Adrian J. Matthews, K. Vijay Kumar, Alejandra Sanchez-Franks, V.
1008　　　Thushara, Jenson George, V. Vijith, Benjamin G. M. Webber, Bastien Y. Queste,
1009　　　Rajdeep Roy, Amit Sarkar, Dariusz B. Baranowski, G. S. Bhat, Nicholas P. Klingaman,
1010　　　Simon C. Peatman, C. Parida, Karen J. Heywood, Robert Hall, Brian King, Elizabeth
1011　　　C. Kent, Anoop A. Nayak, C. P. Neema, P. Amol, A. Lotliker, A. Kankonkar, D. G.
1012　　　Gracias, S. Vernekar, A. C. D'Souza, G. Valluvan, Shrikant M. Pargaonkar, K. Dinesh,
1013　　　Jack Giddings, and Manoj Joshi. 2018. 'BoBBLE: Ocean–Atmosphere Interaction and
1014　　　Its Impact on the South Asian Monsoon', Bulletin of the American Meteorological
1015　　　Society, 99: 1569-87.
1016　Wadley, M.R., D.P. Stevens, T.D. Jickells, C. Hughes, A.R. Baker, R.J. Chance, H. Hepach,
1017　　　L. Tinel, and L.J. Carpenter. 2020. 'A Global Model for Iodine Speciation in the Upper
1018　　　Ocean', Global Biogeochemical Cycles, *In Press, DOI: 10.1029/2019GB006467*
1019　Waite, T. J., V. W. Truesdale, and J. Olafsson. 2006. 'The distribution of dissolved inorganic
1020　　　iodine in the seas around Iceland', Marine Chemistry, 101: 54-67.
1021　Wang, F., A. Saiz-Lopez, A. S. Mahajan, J. C. Gómez Martín, D. Armstrong, M. Lemes, T.
1022　　　Hay, and C. Prados-Roman. 2014. 'Enhanced production of oxidised mercury over the

1023   tropical Pacific Ocean: a key missing oxidation pathway', Atmospheric Chemistry and
1024   Physics (Print), 14: 1323-35.

1025 Wong, G. T. F. 2001. 'Coupling iodine speciation to primary, regenerated or "new" production:
1026   a re-evaluation', Deep-Sea Research Part I-Oceanographic Research Papers, 48: 1459-
1027   76.

1028 Wong, G.T.F. and Brewer, P.G., 1977. Marine chemistry of iodine in anoxic basins.
1029   Geochimica et Cosmochimica Acta, 41(1): 151-159.

1030 Wong, G.T.F., Brewer, P.G. and Spencer, D.W., 1976. Distribution of particulate iodine in the
1031   Atlantic Ocean. Earth and Planetary Science Letters, 32(2): 441-450.

1032 Wong, G. T. F., and X. H. Cheng. 1998. 'Dissolved organic iodine in marine waters:
1033   Determination, occurrence and analytical implications', Marine Chemistry, 59: 271-81.

1034 Wong, G. T. F., and L. S. Zhang. 2003. 'Geochemical dynamics of iodine in marginal seas: the
1035   southern East China Sea', Deep-Sea Research Part II-Topical Studies in Oceanography,
1036   50: 1147-62.

1037 Zic, V., M. Caric, and I. Ciglenecki. 2013. 'The impact of natural water column mixing on
1038   iodine and nutrient speciation in a eutrophic anchialine pond (Rogoznica Lake,
1039   Croatia)', Estuarine Coastal and Shelf Science, 133: 260-72.

1040