Supplementary Material for 'Estimation of Reactive Inorganic Iodine Fluxes in the Indian and Southern Ocean Marine Boundary Layer'

Swaleha Inamdar[1,2], Liselotte Tinel[3], Rosie Chance[3], Lucy Carpenter[3], P. Sabu[4], Racheal Chacko[4], Sarat C. Tripathy[4], Anvita U. Kerkar[4], Alok K. Sinha[4], P. V. Bhaskar[4], Amit Sarkar[4,5], Rajdeep Roy[6], Tomas Sherwen[3,7], Carlos Cuevas[8], Alfonso Saiz-Lopez[8], Kirpa Ram[2] and Anoop S. Mahajan[1]*

[1]Centre for Climate Change Research, Indian Institute of Tropical Meteorology, Dr Homi Bhabha Road, Pashan, Pune, 411 008, India

[2]Institute of Environment and Sustainable Development, Banaras Hindu University, Varanasi, 221005, India

[3]Wolfson Atmospheric Chemistry Laboratories, Department of Chemistry, University of York, YO10 5DD, UK

[4]National Centre for Polar and Ocean Research, Goa, 403 804, India

[5]Environment and Life Sciences Research Centre, Kuwait Institute for Scientific Research Centre, Al-Jaheth Street, Shuwaikh, 13109, Kuwait

[6]National Remote Sensing Centre, Department of Space Government of India Balanagar, Hyderabad, 500 037, India

[7]National Centre for Atmospheric Science, University of York, York YO10 5DD, UK

[8]Department of Atmospheric Chemistry and Climate, Institute of Physical Chemistry Rocasolano, CSIC, Madrid, Spain.

* corresponding author: Anoop S. Mahajan (anoop@tropmet.res.in); phone: +91 20 2590 4526

Supplementary Text

1. Abbreviations used in the text:

ISOE-8	8th Indian Southern Ocean Expedition
IIOE-2	2nd International Indian Ocean Expedition
ISOE-9	9th Indian Southern Ocean Expedition
SK-333	Sagar Kanya-333 expedition in the south Indian Ocean
BoBBLE	Bay of Bengal Boundary Layer Experiments
Chl-*a*	Chlorophyll-*a*
HYSPLIT	HYbrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model (Rolph et al., 2017; Stein et al., 2015)
DSCD(s)	Differential slant column density(ies)

2. Multiple linear regression analysis

A region-specific parameterisation tool for estimating sea surface iodide concentration was developed following the (Chance et al., 2014) method. Observations for SSI concentrations from ISOE-9, SK-333 and BoBBLE were used for the multiple linear regression analysis against various atmospheric and oceanic parameters. SST data used for linear regression analysis was recorded during ISOE-9 using a bucket thermometer at an interval of 6 hours accounting for a change of approximately 1 degree in the latitudinal track of the ship and simultaneously during each seawater iodide sampling. Seawater samples collected during ISOE-9 at the same interval as the recorded SST were used for salinity retrievals using the AUTOSAL salinometer. For the SK-333 and BoBBLE expeditions most of the samples were collected from the CTD casts and some (4 data points) were underway samples in case of

BoBBLE expedition. The MLD climatological monthly mean computed from climatological monthly mean profiles of potential temperature and potential density based on three different criteria was obtained using the World Ocean Atlas (Monterey and Levitus, 1997). That being; a temperature change from the ocean surface of 0.5 degree Celsius (MLD_{pt}), a density change from the ocean surface of 0.125 (sigma units) (MLD_{pd}), and a variable density change from the ocean surface corresponding to a temperature change of 0.5 degree Celsius (MLD_{vd}) (Monterey and Levitus, 1997). All three climatological monthly mean MLD data types (MLD_{pd}, MLD_{pt}, and MLD_{vd}) were used for linear regression with measured iodide concentration. The climatological monthly mean of sea surface nitrate concentrations for ISOE-9 was also obtained from the World Ocean Atlas 2013, version 2 (Garcia et al., 2013). Chl-*a* data for ISOE-9 was obtained from pigment analysis during ISOE-9.

3. Dataset used for SSI estimation

For estimating the SSI concentrations for all campaigns (ISOE-8, IIOE-2, and ISOE-9) using parameterisation methods in Eq. (1) to (5), the oceanic parameters were obtained from the observations during individual campaigns. Missing data was substituted with available data, as in the case of IIOE-2 campaign. For this campaign, salinity data was obtained from the World Ocean Atlas 2013, version 2 (Zweng et al., 2013). Also, chl-*a* data for IIOE-2 was obtained from level 3 daily and 8-day products of Aqua MODIS satellite (NASA-GSFC, 2017). Similarly, sea surface nitrate concentrations and MLD for all the campaigns were obtained from World Ocean Atlas (Garcia et al., 2013; Monterey and Levitus, 1997).

4. Parameterisation for SSI estimation

Chance et al. (2014) developed two versions of empirical relationship for SSI estimation. The first one is given in the main text (Eq. 1). For the regional specific modification (Eq. 2 and 3), each of the oceanic parameters was obtained for the same location (SST, salinity, chl-*a*) as the measured SSI concentrations from ISOE-9, SK-333 and BoBBLE. Likewise, monthly climatological datasets were obtained for MLD and nitrate with a one-degree spatial resolution, as described in the previous section. The regression analysis for region-specific modification was initially divided in three sections – first for the all the SSI observations including the Indian Ocean and the Southern Ocean (ISOE-9, SK-333, and BoBBLE). The second only for the Southern Ocean region (ISOE-9) and lastly only the Indian Ocean region (SK-333 and BoBBLE). The third scenario was rejected due to poor and insignificant coefficient of determination values (R^2) for individual parameters. The resulting parameterisation too was unable to fit the observations with predicted values for the Indian Ocean region. In this case, SST and latitude were the only parameters that correlated positively to the SSI. The first and second scenario resulting in parametrisation denoted by Eq. (2) and Eq. (3) respectively is given in the main text. A list of R^2, slope, intercept and significance of all parameters for linear regression with observed iodide concentration is provided in Table S1. A combination similar to the Chance parameterisation given in Eq. (2) gave maximum R^2 value of 0.794 (N = 128) for the Indian Ocean and the Southern Ocean region. In this equation, all parameters are significant except for salinity and nitrate concentration. Removal of any one of these insignificant parameters did not make the other significant. The coefficient for this equation (Eq. 2) also remained insignificant with high error value (22 ± 137). The combination of SST^2, latitude, nitrate and salinity resulted in a maximum $R^2 = 0.86$ (N=110) for the dependent variable [iodide] in Eq. (3). The inclusion of MLD_{pt} (with highest R^2 for MLD) increased the R^2 slightly but had a non-uniform distribution of the residuals and was thus excluded. Similarly, the addition of chl-*a* to the equation did not change the R^2 significantly, and thus chl-*a* was removed from the final equation. SSI concentration estimated using the logarithmic parameterisation by Chance et al. (2014) was very high in comparison to the measured SSI concentration from ISOE-9. The ln[iodide] equation estimated SSI concentrations of ~500 nM in the Indian Ocean region which is very high compared to global observations of SSI in the Indian Ocean (Chance et al., 2014, Chance et al., 2019) and in comparison to the observations from SK-333 and BoBBLE for the South Indian Ocean. Therefore, we excluded the logarithmic parametrization for this study and suggest that the ln[iodide] parametrization is not adequate for SSI estimation.

5. References

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6. Figures

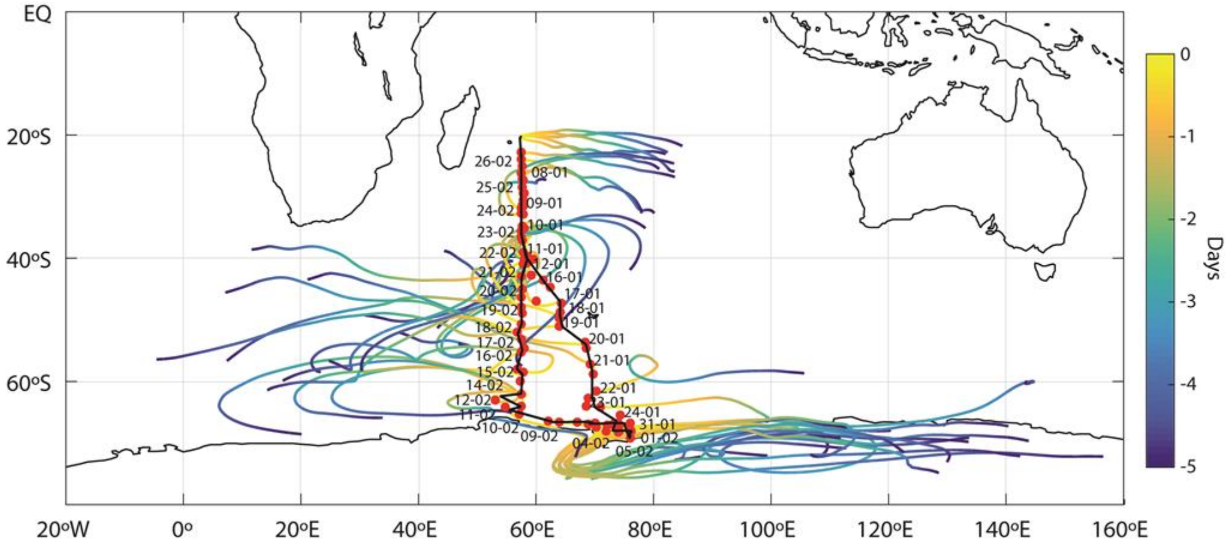

Figure S1: Map of the south Indian Ocean and the Southern Ocean showing the cruise track (black line) for the ISOE-9 campaign. Along the cruise track 5-days backward wind trajectories (HYSPLIT) of the air masses arriving the locations at noon each day of the ISOE-9 expedition. Sea surface iodide sampling locations marked in red circles along with the date of sampling.

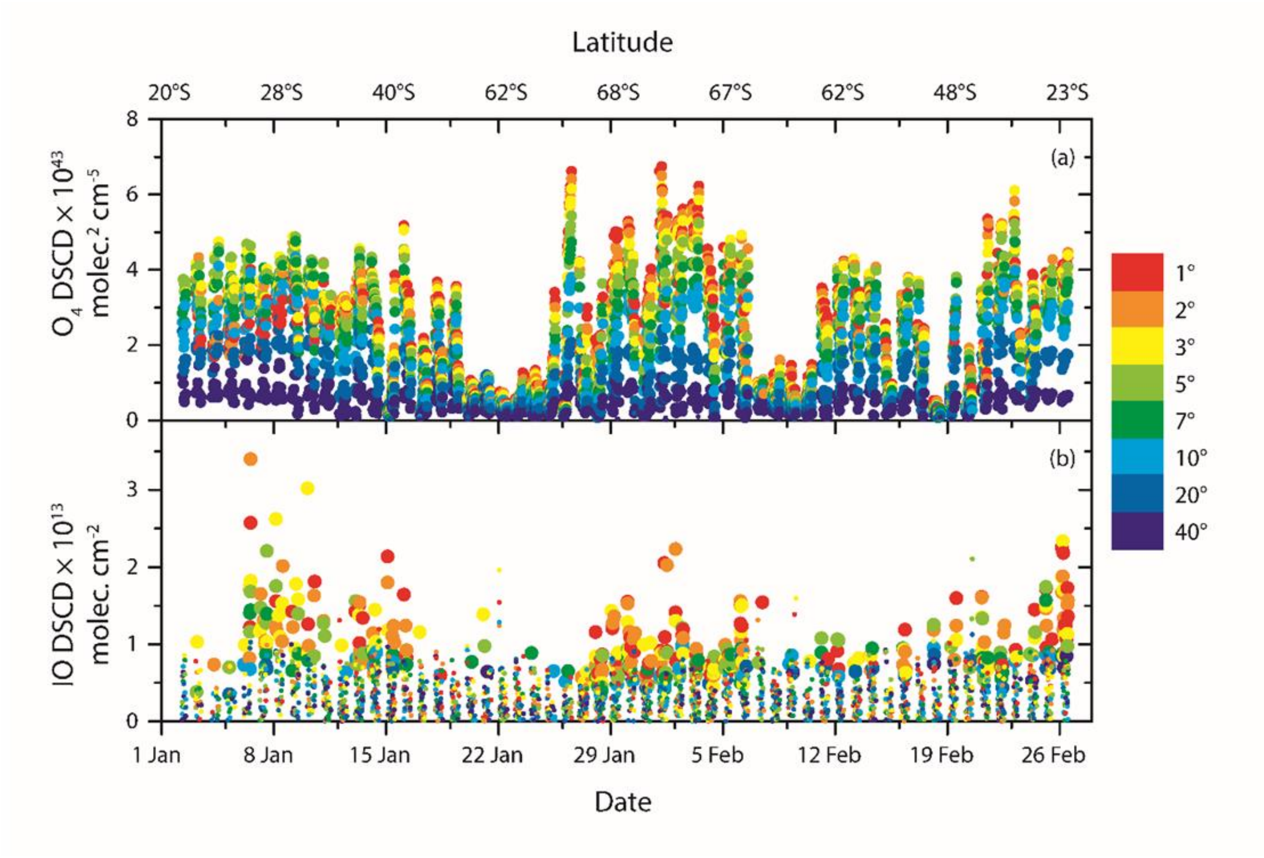

Figure S2: Timeline of the O_4 and IO DSCDs observed during the ISOE-9 expedition. The top scale indicates corresponding latitudes for the dates, and colour code represents the elevation angle (°) for each scan. Smaller circles indicate DSCDs below σ detection limit for IO and 2σ in case of O_4; bigger circles indicate DSCDs above the detection limit respectively.

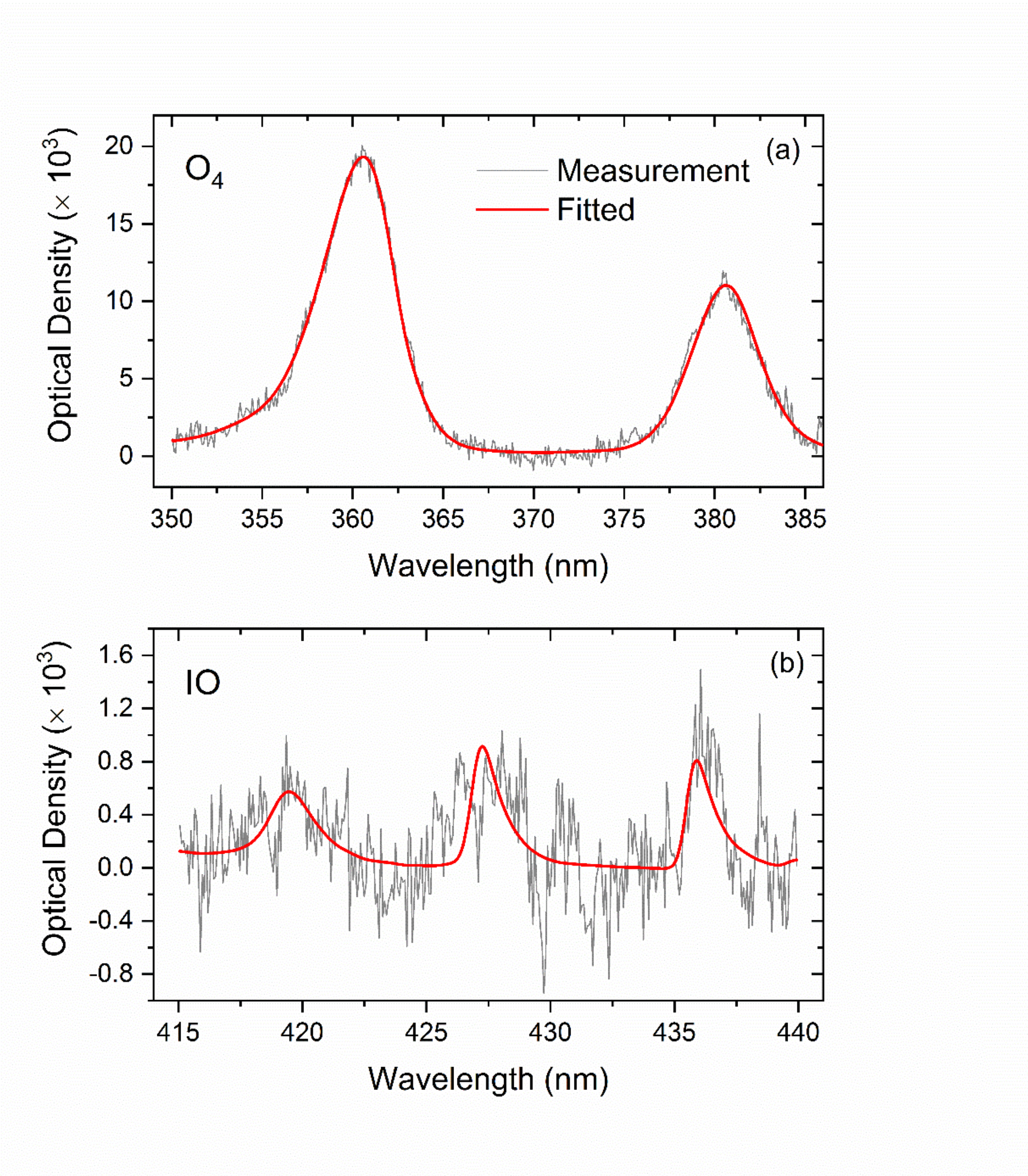

Figure S3: An example of typical spectral fit for O_4 (a) and IO (b) during the ISOE-9 expedition. These spectral fits were taken on 26 February 2017 at 15:35 (local time), for solar zenith angle 69.5° and 1° elevation angle. These fits retrieved O_4 slant column density of $(4.35 \pm 0.035) \times 10^{43}$ molecules cm^{-2} and $(2.24 \pm 0.36) \times 10^{13}$ molecules cm^{-2} with residual optical density (root mean square) of $3.2 \times 10^{-4}$and 5.5×10^{-4} respectively.

7. Tables

Parameter	R^2	Slope (m)	Intercept (C)	p < 5%? (p)
SST	0.64	4.26 ± 0.29	31 ± 4.77	Yes (0)
	0.62	*4.03 ± 0.304*	*32.2 ± 4.17*	*Yes (0)*
1/SST (K^{-1})	0.62	-345781 ± 23910	1297 ± 83.9	Yes (0)
	0.59	*-322918 ± 25302*	*1215 ± 89.5*	*Yes (0)*
SST^2	0.73	0.16 ± 0.0085	41.1 ± 3.6	Yes (0)
	0.79	*0.18 ± 0.01*	*39.2 ± 2.7*	*Yes (0)*
NO3	0.42	-3.24 ± 0.34	125 ± 5.7	Yes (0)
	0.39	*-2.63 ± 0.32*	*110.6 ± 5.8*	*Yes (3.06×10^{-13})*
|Latitude|	0.55	-2.1 ± 0.17	178.3 ± 8.3	Yes (0)
	0.52	*-2.43 ± 0.22*	*196.1 ± 11.7*	*Yes (0)*
Monthly MLD_{pt}	0.17	-1.1 ± 0.22	125 ± 9.2	Yes (1.2×10^{-6})
	0.08	*-0.63 ± 0.21*	*97.6 ± 9.4*	*Yes (0.003)*
Monthly MLD_{vd}	0.04	-0.48 ± 0.2	98 ± 8	Yes (0.03)
	0.003	*-0.11 ± 0.19*	*75.9 ± 7.5*	*No (0.56)*
Monthly MLD_{pd}	0.12	-0.67 ± 0.16	110 ± 7.8	Yes (5.2×10^{-5})
	0.05	*-0.35 ± 0.15*	*87.1 ± 7.7*	*Yes (0.02)*
Salinity	0.08	16 ± 4.8	-468 ± 165	Yes (0.001)
	0.23	*21.8 ± 3.8*	*-675 ± 130*	*Yes (8×10^{-8})*
Chlorophyll - *a*	0.025	-37 ± 26	84 ± 8.6	No (0.16)
	0.002	*-7 ± 20*	*62 ± 7*	*No (0.73)*

Table S1: Linear regression analysis results for each parameter against field observations of sea surface iodide for paramterisation Eq. (2) in standard font and Eq. (3) in italics. R^2 represents the coefficient of determination (COD); the last column is a check for statistical significance at 5% with the p-value in parenthesis.