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### A seasonal OH minimum region over the Indian Ocean?

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# 48 HIGHLIGHTS

- 49 There exists a seasonal OH minimum region over Indian Ocean in spring
- 50 A secondary minimum in autumn, but values are slightly higher than that in spring
- 51 SO<sub>2</sub>, CO, O<sub>3</sub> and H<sub>2</sub>O chemistry play a key role in the observed minimum in the region
- 52 The OH minimum can affect the air quality in the Indian Ocean region

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### 72 ABSTRACT

73 As the hydroxyl radical (OH) is the cleansing agent of the atmosphere, reduction in its concentration is a 74 great concern for air quality and transport of trace gases across the latitudes and altitudes. In addition, OH 75 determines the lifetime of most trace gases and non-CO<sub>2</sub> greenhouse gases in the atmosphere. Since many 76 pollutants have adverse health effects and are greenhouse gases, the changes in OH concentrations directly 77 or indirectly affect public health and climate. Our analysis with OH data (from Tropospheric Emission 78 Spectrometer and Copernicus Atmosphere Monitoring Service reanalyses) for the past 14 (2005–2018) 79 years finds an OH minimum region over Indian Ocean, in the eastern Bay of Bengal at 6°-14°N and 92°-80 95°E. The Indian Ocean OH minimum reaches to  $15 \times 10^4$  molec.cm<sup>-3</sup> in April, and a secondary minimum of 81 18×10<sup>4</sup> molec.cm<sup>-3</sup> in September–November. This seasonal minimum found around the Andaman and 82 Nicobar Islands also hosts an active volcano, which releases noticeable amount of  $SO_2$  (OH sink) throughout 83 the year. In addition, the biomass burning in spring and thus, the distribution of CO has a profound influence 84 on the OH distribution in this region as Southeast Asia is one of the global hotspots of biomass burning, and 85 the Indian Ocean OH minimum is located near that region. The El Niño and La Niño events also control the 86 tropospheric ozone and CO distribution, and thus the interannual variability of OH minimum there. The 87 WACCM model simulations reproduce the general latitudinal distribution and average seasonal cycle of 88 OH, but not the extreme minimum OH values, although the simulations show the annual minimum in winter 89 (December–January) in both ocean regions, which demands dedicated studies using chemical transport 90 models. Therefore, this study reveals a seasonal OH minimum over Indian Ocean, which is very likely to 91 influence the regional air quality and trace gas transport in the tropics.

92

93 Short title: Indian Ocean OH minimum

94 Key words: Indian Ocean; Stratosphere; troposphere; Pollution; western Pacific Ocean; OH

96 1. Introduction

97 Hydroxyl (OH) is a short-lived (less than a second) free radical and the most effective scavenger in the 98 troposphere. It is the major oxidant in the lower troposphere that determines the lifetime of most trace 99 gases and greenhouse gases (GHGs) such as methane (CH<sub>4</sub>) in the atmosphere (Lelieveld et al., 2016; Nicely 100 et al., 2018), and therefore, is a climate relevant gas. OH radical is called the "detergent of atmosphere" 101 due to its role and ability to clean the atmosphere. The general composition of stratosphere is influenced 102 by the species that are transported from the troposphere. Hydroxyl radicals in the troposphere react with 103 some short-lived species and convert them into soluble products. Therefore, OH radicals in the troposphere 104 mask the stratosphere from many tropospheric emissions (Rex et al., 2014). The variability of OH in the 105 troposphere is key not just for the supply of chemical species such as SO<sub>2</sub> and CH<sub>4</sub> to the stratosphere, but 106 also for the atmospheric lifetime of several trace gases (Manning et al., 2005).

107

There are multitude of sources and sinks for the hydroxyl radical. The primary production of OH is by the
photodissociation of ozone by ultraviolet (UV) sunlight with a wavelength of the photon (hv) shorter than
330 nm in the presence of water vapor (Levy, 1971):

111 $O_3 + hv (\lambda < 330 \text{ nm}) \rightarrow O(^1D) + O_2$	(R 1)
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**112**  $O(^{1}D) + H_{2}O \rightarrow 2OH$  (R 2)

Recently, it is found that this source has a relatively small contribution compared to other sources under
polluted conditions (e.g. Elshorbany et al., 2010; Vogel et al., 2003). Photolysis of HCHO is a daytime source
of HO<sub>2</sub> radicals:

116 HCHO + hv (threshold,  $\lambda < 338 \text{ nm}$ )  $\rightarrow$  H + HCO (R 3)

117  $H + O_2 + M \rightarrow HO_2 + M$  (R 4)

118  $HCO + O_2 \rightarrow HO_2 + CO$  (R 5)

**119** The conversion of  $HO_2$  to OH depends on the availability of NO.

120 $HO_2 + NO \rightarrow OH + NO_2$	(R 6	)
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121 The OH radical is also formed by the ozonolysis of alkenes. In addition, nitrous acid (HONO) contributes to

122 the formation of OH radical:

123 HONO + hv (threshold,  $\lambda < 400 \text{ nm}$ )  $\rightarrow$  OH + NO (R 7)

This is a major source of daytime OH concentration (Elshorbany et al., 2010). The OH radical reacts with
reduced and partly oxidised gases such as CH<sub>4</sub>, CO and non-methane Volatile Organic Compounds
(NMVOC). CO and CH<sub>4</sub> are the direct sinks of atmospheric OH (Lu et al., 1993).

- 127  $CO + OH \rightarrow CO_2 + H$  (R 8)
- 128  $CH_4 + OH \rightarrow CH_3 + H_2O$  (R 9)

129Also, under high NOx conditions, permanent loss processes are dominated by the reaction of OH with NO2.130 $OH + NO_2 (M) \rightarrow HNO_3 (M)$ (R 10)

**131** Therefore, the regional variability of OH depends on several factors.

132

133 An OH minimum has been found in the tropical western Pacific centered at 17° N /158° E. This "OH hole" 134 was reported based on the observations of very small values of tropospheric O<sub>3</sub> in this region (Rex et al., 135 2014). The explanation for this is that suppression of primary OH production due to low  $O_3$  or deficiency of 136 secondary production due to low NO (Wolfe et al., 2019a,b). It implies that the region has very low 137 atmospheric pollution or pristine air. Here, we examine whether there is any OH minimum zone over other 138 ocean basins, e.g. Indian Ocean. Our analyses expose an OH minimum (at the surface level) over Indian 139 Ocean at around 12° N/93° E. Therefore, we present a detailed assessment of the spatio-temporal 140 variability of this OH minimum for the past 14 years (2005–2018) using the TES (Tropospheric Emission 141 Spectrometer) reanalysis data. Since the analysis and identification of a minimum value is very sensitive 142 and could be also data specific, we have used another reanalysis data, the Copernicus Atmosphere 143 Monitoring Service (CAMS). Furthermore, we have also used a global chemical transport model (CTM) for

the simulation of OH in both oceanic regions for examining the representation of OH minimum in the stateof-the-art CTMs. The distribution of OH is compared to that of CO, CH<sub>4</sub>, HCHO, SO<sub>2</sub>, H<sub>2</sub>O, NO<sub>x</sub>, HNO<sub>3</sub> and O<sub>3</sub>
in the minimum region. Note that there are no measurements available in this region, and therefore, we
have used the reanalysed data and model simulations. The influence of meteorological and oceanic factors
such as clouds, water vapour, precipitation, winds and sea surface temperature (SST) on the variability of
OH is also discussed. The OH minimum over this region is compared to that in the tropical western Pacific
and reasons for these minima are assessed.

151

### 152 2. Data and Methods

153 The TES chemical reanalysis data have a horizontal resolution of 1.1°×1.1° (e.g. Cady-Pereira et al., 2017; 154 Miyazaki and Bowman, 2017). Chemical reanalysis is a technique in which measurements from multiple 155 satellites are combined to get a continuous data of atmospheric composition. These data are made by 156 assimilating the measurements from OMI (Ozone Monitoring Instrument), MLS (Microwave Limb Sounder, 157 Livesey et al., 2020), TES (Miyazaki and Bowman, 2017) and MOPITT (Measurements of Pollution in the 158 Troposphere, Deeter et al., 2003) satellite measurements. The reanalysis is made using a global CTM, and 159 an ensemble Kalman filter that optimises the concentrations of different species (Miyazaki et al., 2015). We 160 have used  $O_3$ , CH<sub>4</sub>, NO<sub>x</sub>, HCHO, SO<sub>2</sub>, CO, HNO<sub>3</sub> and OH data from the TES reanalysis.

161

162 CAMS is the global atmospheric composition reanalysis made by the European Centre for Medium-Range 163 Weather Forecasting (ECMWF). It provides 3-dimensional time-consistent quantifications of 164 meteorological variables and concentrations of aerosols, dust and other trace gases such as OH from 2003 165 onwards. The reanalysis product offers a gridded data with a spatial resolution of 0.75°×0.75°. It has 166 relatively smaller biases with respect to the measurements, and is more reliable than that of Monitoring 167 Atmospheric Composition and Climate (MACC), the previous version of CAMS (Inness et al., 2013). All variables in the CAMS reanalysis are assimilated using the IFS Cycle 42r1 4D-Var. Here, we use the monthly
mean OH data at the surface for the Indian and Pacific Oceans. Further information about the data is
provided in Inness et al. (2019).

171

172 To analyse the impact of meteorological factors on OH distribution, we have used the Tropical Rainfall 173 Measuring Mission (TRMM) daily rainfall data with a resolution of 0.25°×0.25° (TRMM, 2011; Kuttippurath 174 et al., 2021). This dataset contains the output of TRMM Algorithm 3B42 to produce the merged TRMM 175 precipitation measurements. The NOAA (National Oceanic Atmospheric Administration) Extended 176 Reconstructed Sea Surface Temperature (ERSST) Version 4 data are also considered (e.g. Huang et al., 2017; 177 Anandh et al., 2018). These monthly mean data have a spatial coverage of  $2^{\circ} \times 2^{\circ}$ . The National Centers for 178 Environmental Prediction (NCEP) reanalysis monthly data are considered for wind, which have 17 pressure 179 levels on a horizontal resolution of 2.5°×2.5°.

180

181 The NASA-EOS Terra and Aqua spacecrafts are equipped with the MODIS sensor. Terra crosses the equator 182 southward around 10:30 local solar time (LST), while Agua crosses northward around 13:30 LST. MODIS 183 observes the Earth in 36 channels in a spectral resolution from 0.41 to 14  $\mu$ m. MODIS uses infrared bands 184 to measure cloud properties in relation to cloud top temperature and pressure. It employs visible and near 185 infrared channels to derive optical and microphysical cloud parameters (King et al., 2003). We use the 186 MODIS level 3 gridded monthly mean cloud fraction data with a spatial resolution of 1°×1° over the Indian 187 Ocean region for the year 2005. The monthly mean data are taken to estimate the seasonal distribution of 188 cloud fraction. We also use the MODIS monthly active fire product at 1 km spatial resolution. The MODIS 189 Adaptive Processing System (MODAPS) real-time active fire data are also available through Fire Information 190 for Resource Management (FIRM). These data are processed by MODAPS to the collection 5 Active fire

- product using an enhanced contextual fire detection algorithm (Giglio et al., 2003). Here, we examine the
  monthly mean active fire data from 2005 to 2018 in the Southeast Asian region adjacent to Indian Ocean.
- 193
- In addition, we have also used the WACCM model simulations to examine the OH minimum over both oceanic basins. This is a three-dimensional offline CTM with full tropospheric chemistry. The model is driven by meteorological data from ECMWF reanalysis (Dee et al., 2011). The model has a horizontal resolution of 1°×1.25°, and the vertical co-ordinates range from the surface to 5×10<sup>-6</sup> hPa. The model results are provided for every 6 h. Further details about the model can be found in Wu et al. (2021) and Marsh et al. (2013).
- 199

For studying the monthly variations in the distribution of OH and other trace gases, their average for each
month from 2005 to 2018 is estimated. Seasonal averages are computed by finding the mean over the
months DJF (December, January and February) for winter, MAM (March, April and May) for spring, JJA
(June, July and August) for summer and SON (September, October and November) for autumn.

204

### 205 3. Results and discussion

### 206 *3.1 The OH minimum in Indian Ocean*

207 Fig. 1 (top) shows the global surface OH distribution, and Fig. 1 (bottom) zooms into the tropical western 208 Pacific (left) where the OH hole (smaller than 1 molec./cm<sup>3</sup>) exists and the adjacent Indian Ocean (right) 209 regions. The OH hole in the western Pacific has already been identified (Rex et al., 2014). However, similar 210 values of OH in other tropical ocean regions are not reported and are rare, although OH concentrations 211 generally decrease with latitude from tropics to the high latitudes. The OH concentrations are largest in the 212 tropical latitudes and along the shipping routes (e.g. Gopikrishnan and Kuttippurath, 2021), and they 213 gradually decrease from 30°-40° N/S to the polar regions. Since the tropical upwelling regions transport 214 atmospheric trace gases to the middle latitudes of stratosphere, the changes in OH concentrations in these latitudes are very important. Fig. 1 (bottom, right) shows an OH minimum in Indian Ocean located at 6°–
14°N and 92°–95°E, which could also be termed as a "remote" extension of the western Pacific OH hole as
the Indian Ocean minimum region is in its vicinity. High reactivity of OH, about a second, is the reason for
its short life span, which makes OH highly variable in time and space. Changes in the concentration of OH
depend on various environmental factors such as humidity and cloudiness, intensity of incoming radiation
and the presence of natural or anthropogenic pollutants (Lelieveld et al., 2004).

221

222 The monthly mean OH analysed for the past 14 years (2005–2018) is shown in Fig. 2 (top). The analysis 223 indicates that the minimum values are spreading towards the west during the months of November, 224 December, January and February. The minimum values are confined to a small region in this period. The 225 OH concentration increases in the western Bay of Bengal (BoB, 8°–18°N, 76°–90°E) afterwards in March 226 and April. The OH minimum confines to a small region in May, June, July and August, and the minimum 227 region (6°–18°N and 80°–100°E) is surrounded by high values (e.g. 150–300×10<sup>4</sup> molec./cm<sup>3</sup>). The trough 228 of minimum is observed at a specific location and it is consistently present in all months, at 6°-14°N and 229 92°–95°E, and the minimum region is found around the Andaman and Nicobar Islands in the northeast 230 Indian Ocean. We analysed another data (CAMS) for the period 2003–2018, which also displayed a very 231 similar monthly cycle (unit of OH concentration is different here) with a region of OH minimum around  $6^{\circ}$ -232 18°N and 80°–100°E, as found in the TES data (Supplemental Fig. 1).

233

Fig. 2 (bottom) shows the monthly OH minimum in the selected latitude-longitude region (6°–18°N and 80°–100°E) averaged for the period 2005–2018. It shows two minima in a year, with the smallest values in April ( $15 \times 10^4$  molec./cm<sup>3</sup>) and a secondary minimum during September–November ( $18 \times 10^4$  molec./cm<sup>3</sup>), depending on year (here in November). Relatively higher values are observed in other months, about 20– 40×10<sup>4</sup> molec./cm<sup>3</sup>. We also analysed the CAMS data to investigate the seasonal distribution of OH minimum, as shown in Fig. 2. The results show very similar monthly progression with the annual minimum in April (0.012 ppt) and the secondary minimum in September–November (0.015 ppt), as found in the TES reanalysis. It suggests that, although the OH minimum zone is vast and spread across hundreds of kilometers, the lowest values of OH appears in April. Henceforth, the OH minimum in Indian Ocean is seasonal and is in a relatively small region as compared to that in the western Pacific. This situation demands a detailed analysis of OH distribution in different seasons over Indian Ocean.

245

### 246 3.2 Seasonal Variability of OH minimum

247 Fig. 3 displays the minimum values of OH in the latitude region 4°–20° N for different seasons. Although 248 the seasonal change is very small in the selected region, comparatively lower values are found in spring 249 (MAM, about 15×10<sup>4</sup> molec./cm<sup>3</sup>), consistent with our previous discussion. On the other hand, the basin-250 wide lowest values (<  $100 \times 10^4$  molec./cm<sup>3</sup>) are found in winter (DJF) and autumn (SON), but relatively 251 higher values (> 200×10<sup>4</sup> molec./cm<sup>3</sup>) outside the minimum region are found in summer and spring, 252 particularly in the western BoB. We also examined the longitudinal distribution of OH in the selected region 253 (Fig. 3, top) and it shows the smallest values  $(20 \times 10^4 \text{ molec./cm}^3)$  at  $92^\circ$ – $95^\circ$  E, which coincides the 254 selected OH minimum region, where the seasonal differences are very small. However, beyond this 255 minimum region (92°–95° E), the lowest OH values are found in winter and autumn. The CAMS data also 256 yield a similar seasonal distribution of OH in BoB and in the selected region (Supplemental Fig. 2). In fact, 257 the most critical element of OH distribution is its diurnal variation (Prinn et al., 1995). It quickly responds 258 to solar radiation and reaches its peak concentration at noon, as the photolysis of  $O_3$  to  $O(^1D)$  depends on 259 solar radiation, and the reaction between  $O(^{1}D)$  and  $H_{2}O$  is the key source for OH (Rohrer and Berresheim, 260 2006). Additionally, the diurnal variation (TES data presented are monthly) depends on seasons, as the 261 intensity of solar radiation at noon is stronger and duration of day is longer in summer. Accordingly, 262 concentrations of OH are higher in summer than that in winter (Lu et al., 1991), as found in our analyses.

263 Fig. 4 displays OH (from CAMS), precipitation, SST, cloud cover,  $SO_2$ , CO and  $NO_X$  for different seasons in 264 2005. In winter, there are lower SST and less precipitation in the northern BoB, above 8° N, and are 265 relatively higher south of that latitude. The OH production is expected to decrease under these 266 temperature conditions and consequently, its concentration is relatively lower during this season. In April, 267 the temperature and SST increase (highest SST in the annual cycle) and thus, higher values of OH can be 268 found in the western BoB, but the annual minimum is observed during this period in the eastern BoB. 269 Furthermore, the winds make a circular pattern around the minimum region north of 6° N. In summer, 270 heavy rainfall is observed at the east coast of BoB, about 20-30 mm. SST is also relatively high in the 271 northern and southeastern BoB. Therefore, OH shows the highest annual concentrations over the basin in 272 this period. The largest OH concentrations are found in the lowest precipitation regions such as 6° N along 273 the meridional transect of BoB, and thus, the OH minimum region shrinks to its smallest in the annual cycle 274 during this period. In autumn, SST of these regions is relatively small, providing a condition for 275 correspondingly large OH minimum zone there.

276

277 Reaction of OH radical in cloud water play an important role in the formation of secondary aerosols. Strong 278 seasonal cycle is observed in cloud fraction over Indian Ocean, where very high values are observed in 279 summer and autumn (0.8–1), but relatively small fraction of clouds in winter and spring (< 0.5). Similar 280 seasonal distribution is also observed in the region of OH minimum, where the highest cloud fractions are 281 found in summer and autumn, and smallest in winter. However, the analysis shows that the impact of 282 clouds on the smallest OH values in the minimum region is limited.

283

Since there is an active volcano in the Andaman Islands, we also examined the concentration of  $SO_2$  in the region as it is a sink for atmospheric  $SO_2$  (third panel from bottom). In general, relatively large concentrations are observed in the Indian coastal regions, where the seasonal highest values are observed 287 in spring  $(3-4 \,\mu g/m^3)$ . The values are very small over the open ocean, but relatively high SO<sub>2</sub> concentrations 288 are found in the OH minimum region, which is present throughout the year. The seasonal differences are 289 very small and therefore, there are other factors controlling the OH minimum in this region. Biomass 290 burning produces a large amount of CO in earth's atmosphere, mostly within 2 km. Lu et al. (1993) showed 291 that the decrease of CO implies an increase of OH (Reaction R8). In general, the amount of CO is high during 292 winter, which decreases as summer arrives. The highest concentration of CO is observed in winter and then 293 in autumn, and the smallest in summer. Furthermore, ozone is the primary precursor of OH radical and 294 therefore, the changes in ozone decides the distribution of OH in the atmosphere. The ozone distribution 295 also follows the distribution of CO, as it shows basin-wide high concentration in winter and autumn, and 296 low values in summer and spring (Supplemental Fig. 3). The minimum ozone is also found in the same 297 region of OH minimum in spring; consistent with that found in the western Pacific (Rex et al., 2014). 298 However, there are still some other processes that could drive the seasonal OH minimum in Indian Ocean, 299 as the meteorology or ocean state are not enough to explain this. Therefore, we explore the changes in 300 tropospheric chemistry to find the additional causative factors.

301

302 Being the major oxidant in the troposphere, the distribution of OH radical is connected to several trace 303 gases in the atmosphere. We analysed the monthly average of OH and its source/sink trace gases in the 304 selected region and are displayed in Fig. 5. The OH distribution shows its highest values in summer (June, 305 July and August), which decrease gradually with time until April. The smallest values are observed in winter 306 and spring (December through April). The fire counts and CO distribution follow similar temporal 307 progression until September, as biomass burning is a major source for CO, and OH shows smaller values 308 during the periods of high CO concentrations. Similarly, one of the mechanisms by which OH is lost is its 309 reaction with NO<sub>2</sub> under high NO<sub>x</sub> conditions. The formation of HNO<sub>3</sub> normally indicates the reaction of OH 310 with  $NO_2$  and the loss of OH (i.e., R10); a reason for the opposite monthly distribution of  $NO_2$  and OH here.

311 Besides, the distribution of OH radical is associated with certain other trace gases such as HONO,  $CH_4$  and 312 HCHO, and changes in the concentration of those could also make analogous variation in OH distribution, 313 as illustrated in the figure. The trace gases that are the sink of OH show an opposite distribution to that of 314 OH, as illustrated by CH<sub>4</sub>, CO and SO<sub>2</sub>, but show the lowest values in April during the annual OH minimum. 315 Seasonal variability of OH depends on a number of factors, such as the changes in solar radiation and the 316 amount of water vapor, as H<sub>2</sub>O has a major role in the production of OH radicals (e.g. see the temporal 317 distribution of  $H_2O$  in the figure). The primary production of OH by photolysis of ozone happens in the 318 presence of water vapour. In addition, photolysis of H<sub>2</sub>O at wavelength in first continuum is an alternate 319 source for the production of OH radical (Ung and Back, 1964): 320  $H_2O + hv \rightarrow H + OH$ (R 11) 321 As shown by Lu et al. (1991), in an unpolluted troposphere, 50% of OH is generated by the reaction between 322  $H_2O$  and  $O(^1D)$  as the direct source, as demonstrated in the figure. 323

#### 324 3.3 Inter-annual variability of OH minima

325 Interannual variability of global OH concentration is mainly connected to the El Niño Southern Oscillation 326 (ENSO) events. Periods of La Niña are positively correlated with an increase in OH. This correlation is due 327 to the increased occurrence of lightning emission of NO<sub>x</sub> during La Niña owing to more convective activity 328 during the period (Turner et. al., 2018). Biomass burning related CO emissions is another event, which may 329 also trigger the variability of OH in the atmosphere. Since Southeast Asia is a zone of biomass burning, the 330 interannual variability of OH can be highly influenced by these fire events. In general, biomass burning is 331 episodically restricted and it peaks during March-April in Southeast Asia (Huang et al., 2013; Kuttippurath 332 et al., 2020), as exhibited in the fire counts data in Fig. 5.

334 Fig. 6 shows the interannual variability of OH in its minimum zone for the period 2005–2018. The spatial 335 variation of OH (averaged values are shown in the top panel and minimum values in the second panel from 336 the top; averaged over 90°–98°E) for different years are depicted. The black contours indicate  $20 \times 10^4$ 337 molec./cm<sup>3</sup>. The OH climatology over the region shows a consistent pattern of OH minimum in Indian 338 Ocean. The seasonal minimum of OH in spring is clearly visible with concentrations of about  $15 \times 10^4$ 339 molec./cm<sup>3</sup> between 8° and 14° N, and the lowest around 12° N. On the other hand, high OH concentrations 340 are observed at 5°–8° N and north of 14° N (>  $200 \times 10^4$  molec./cm<sup>3</sup>). Since this is the monthly climatology, 341 the interannual changes are hardly visible. Therefore, we also examined the minimum values of each month 342 (second panel from the top). The analyses show the lowest OH concentrations between 10° and 13° N, and 343 are blow  $20 \times 10^4$  molec./cm<sup>3</sup>. The latitude regions north and south of these seasonal minima show OH 344 values of about  $50-100 \times 10^4$  molec./cm<sup>3</sup>. The highest OH values are observed beyond these minima zones, 345 i. e. at 7°–5° N and 17°–19°, about  $200 \times 10^4$  molec./cm<sup>3</sup> or higher. The inter-annual changes are very large 346 and explicit in these analyses. For instance, the minimum values are found in relatively smaller regions in 347 2005, 2011 and 2012.

348

349 To diagnose the characteristics of minimum region and its year-to-year changes, we found the average 350 values in each month for the minimum region and are shown in the bottom four panels. The lowest 351 minimum in the TES data is observed in 2009 and 2016 (around  $60 \times 10^4$  molec./cm<sup>3</sup>), and the yearly minima 352 are in the range of  $40-60 \times 10^4$  molec./cm<sup>3</sup>. In general, the minimum peaks are found in spring. CO emitted 353 from biomass burning might have caused this variability, as this is the peak biomass burning season in South 354 Asia (Huang et al., 2013). Conversely, high values are observed in December and January, and even the 355 minimum trough was absent in 2010. As mentioned earlier, the inter-annual variability of ozone and CO is 356 highly connected to ENSO events (e.g. Xue et al., 2021). Therefore, there is a strong relationship between 357 ENSO events and OH minimum. In most years, we find higher values of OH during La Niña periods. For instance, the year 2010 had a strong La Niña, which could be the reason for the absence of OH minimum
in that year. Since H<sub>2</sub>O is a source for OH, its monthly and yearly distributions are in agreement with those
of H<sub>2</sub>O. Conversely, as CO, SO<sub>2</sub> and CH<sub>4</sub> are known to be the sink of OH, their monthly and yearly
distributions are opposite to those of OH, except in the annual minimum period of April. These analyses
also expose relatively high values of OH from 2015 onwards, which can be due to the slight reduction in its
sink (three bottom panels).

364

### 365 3.4 Comparison with the western Pacific OH minimum

366 An OH minimum or OH hole has already been reported in the tropical western Pacific. Very small values of 367 ozone in this region (i.e. clean air) is the main reason for this (Rex et al., 2014). Tropical pacific is the major 368 source/pathway of trace gases to the stratosphere and thus, the region has a great significance in terms of 369 atmospheric composition. Here, we compare the OH hole in the Pacific with the Indian Ocean OH minimum, 370 as it is adjacent to the Pacific Ocean that plays a key role in stratospheric and tropospheric exchange of 371 trace gases. Fig. 7 shows the OH minimum values in Indian and Pacific Oceans for each month from 2005 372 to 2018. It is evident that the western Pacific OH minimum is small enough to call a "hole" and is present 373 throughout the year. In contrast, the OH minimum in Indian Ocean is seasonal, where the lowest values 374 appear in April. As mentioned, the lowest OH minimum in Indian Ocean is about  $10-20 \times 10^5$  molec./cm<sup>3</sup>, 375 depending on month and year. The CAMS data (dashed lines) replicate the monthly and interannual 376 variability of OH minimum in both oceanic regions, as demonstrated with the TES data.

377

### 378 3.5 Modelled OH minimum

To examine the existence of OH minimum, we simulated the distribution of OH in the Indian and Pacific
Oceans using the WACCM model (Fig. 8). The model reproduces the general features of OH distribution
across the latitudes, with the highest concentrations in the tropics, which decreases with latitude towards

382 the poles. The model shows the highest values in summer (June–September) and the smallest in autumn, 383 winter and spring (in March), as for the TES data. Therefore, the seasonal cycle shows the highest values in 384 summer and spring, and the smallest in winter and autumn, as found with the TES data outside the OH 385 minimum region (Supplemental Fig. 4). However, the model hardly finds the seasonal minimum in the 386 selected region, where the annual minimum is as it is found for the other regions over the basin. The 387 simulations for different years and for the western Pacific (Supplemental Fig. 5) indicate that the model 388 struggles to capture the OH hole, although there is a seasonal minimum in winter as over Indian Ocean. 389 Horizontal resolution of the model can be one of the primary reasons for this difference, although different 390 chemical reactions, representation of photolysis of O<sub>3</sub>, and the modelled results of ozone and CO play a 391 decisive role in simulating OH values by the model (e.g. Nicely et al., 2017). Since OH minima or hole are 392 extreme values, simulating this atmospheric chemistry would be a great challenge for any CTM. Therefore, 393 dedicated model studies are required to infer the simulations of such extreme cases. This is a future study 394 as the model sensitivity assessment is beyond the scope of this work.

395

### 396 *3.6 Limitation of the study*

397 This study is based on the renalysed data as there are no direct measurements from *in situ* instruments or 398 satellites in the region concerned. Since these data are heavily weighted by the simulations from a model 399 that used for making the reanalysis, our assessment with the same also has this particular limitation, 400 because modelling of OH is challenging and many uncertainties are still there (e.g. Nicely et al., 2018). This 401 is the reason that we used two different reanalysed data sets here. In addition, we have also used model 402 results for examining the representation of OH minimum in CTMs. The region in which the OH minimum 403 has found is around the Andaman and Nicobar Islands and there is an active volcano on the island. This 404 suggests that the island land mass also has a role in the particular distribution OH there. Our study reiterate 405 the need of more OH and related trace gas measurements in the region for a solid conclusion about the406 OH minimum over Indian Ocean.

407

408 4. Conclusions

409 The OH distribution over Indian Ocean exhibits two minima, in spring and autumn, as assessed from the 410 TES and CAMS renalyses. The spring concentrations reach to  $15 \times 10^4$  molec./cm<sup>3</sup> and slightly higher in 411 autumn (SON). The minimum region is found near the Andaman and Nicobar Islands in Indian Ocean, where 412 it houses an active volcano. The concentration of SO<sub>2</sub> is relatively higher there, which is also the only region 413 over BoB with high SO<sub>2</sub> concentrations, except near the coasts. Since SO<sub>2</sub> is one of the sinks of OH, this 414 could be a reason for the OH minimum there. In general, the sink trace gases (e.g. CO, SO<sub>2</sub> and CH<sub>4</sub>) show 415 an opposite temporal distribution to that of OH in the minimum region. The minimum troughs were present 416 in all years, except in the strong La Niña year 2010. The OH distribution shows an anti-correlation with ENSO 417 events, and therefore, relatively higher OH concentrations are found during the La Niña years. The analyses 418 also reveal that the biomass burning and CO distribution have significant influence on the distribution of 419 OH minimum, as the OH chemistry is closely linked to the changes in  $O_3$ , CO and HNO<sub>3</sub>. The model struggles 420 to capture the OH minimum in both oceans, which demand more dedicated studies with high resolution 421 simulations. In brief, the OH minimum in the western Pacific Ocean is owing to its clean air there, but it is 422 very likely due to the polluted air over Indian Ocean. As OH has the control on the cleansing capacity of 423 atmosphere, its changes will affect air quality and troposphere-stratosphere exchange, which in turn, would 424 affect public and ecosystem health, and climate of the region. Therefore, this study suggests a continuous 425 monitoring of atmospheric OH over Indian Ocean.

426

427

## 429 Credit authorship contribution statement

430 JK: Conceptualization, Methodology, Supervision, Visualization, Writing-original draft, review & editing of

- 431 the original draft, AD, SR, WF: Methodology, Data Analysis, Visualization, Writing original draft
- 432

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### 443 Data availability

- 444 The data used in this study are publicly available.
- 445 TES chemical reanalyses : https://tes.jpl.nasa.gov/chemical-reanalysis/,
- 446 TRMM on http://mirador.gsfc.nasa.gov/cgibin/mirador/
- 447 ERSST data: <u>https://www1.ncdc.noaa.gov/pub/data/cmb/ersst/v5/netcdf/</u>.
- 448 MERRA 2 data are available on: <u>https://giovanni.gsfc.nasa.gov/giovanni/</u>
- 449 The CAMS data are available on: <u>https://ads.atmosphere.copernicus.eu/#!/search?text=reanalysis</u>
- 450 MODIS fire count data: <u>https://firms.modaps.eosdis.nasa.gov/active\_fire/</u>
- 451

452 A	dditional	Information
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### 453 Declaration

454 The authors confirm that there are no known conflicts of interest associated with this article.

455 Competing Interest

456 The authors declare no competing interest.

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Fig. 2. The monthly distribution of surface OH over the eastern north Indian Ocean for the period 2005–
2018. The regional average of monthly mean surface OH for the minimum region is illustrated in the bottom
panel. The months are represented by their first three letters in the top panel (e.g. JAN is January) and by
their first letter on the x-axis of bottom panel (e.g. J is for January and D is for December).





Fig. 3. Top: distribution of surface OH averaged over the latitude band 4°-20° N (i.e. minimum region) for
different seasons. Bottom: distribution of surface OH averaged for each season for the period 2005-2018
in the eastern north Indian Ocean. The seasons are defined as winter (DJF), spring (MAM), summer (JJA)
and autumn (SON).



Fig. 4. Surface concentration of OH from CAMS, precipitation (winds overlaid), SST, cloud cover, SO<sub>2</sub>, CO
and NO<sub>x</sub> for different seasons in eastern north Indian Ocean for the year 2005. The northern hemispheric
seasons are defined as winter (DJF), spring (MAM), summer (JJAS) and autumn (ON). Here, summer season
includes the month of September to examine the monsoon precipitation.

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Fig. 5. Monthly distribution of surface OH and other trace gases averaged over the OH minimum region
(6°-14°N and 92°-95°E) for the period 2005-2018. The months are represented by their first letter on the
x-axis (e.g. J is for January and D is for December).

- ....



Fig. 6. Surface OH concentration averaged over the longitudes 90°–98°E for the period 2005–2018. Here,
the top panel shows the average of surface OH (top) and the second panel from top shows the minimum
of surface OH in the selected region. The bottom panels show the concentrations of mean surface OH
estimated from the TES and CAMS reanalysis data (third panel from top), and different trace gases averaged
over the minimum region for the study period. All data are monthly averaged.

641





**Fig. 8.** Surface OH values simulated by the WACCM model for different seasons of 2005. The seasons are

657 defined as winter (DJF), spring (MAM), summer (JJA) and autumn (SON).

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