

Decadal variations in natural and anthropogenic aerosol optical depth over the Bay of Bengal: Influence of pollutants from Indo-Gangetic Plain.

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Abstract

Perennial increase in atmospheric pollution over the Bay of Bengal (BoB) and South China Sea is reported due to increase in human population and industrial activity in South and Southeast Asia. Based on total aerosol optical depth (AOD) derived from MODIS (Moderate Resolution Imaging Resolution Imaging Spectroradiometer), natural and anthropogenic fractions were derived. The seasonality and spatial variability in rate of increase in total, natural and anthropogenic AOD fractions were examined over the BoB using data collected between 2001 and 2019. Both total and anthropogenic AOD displayed statistically significant rate of increase in the northwest (NWB) and western coastal BoB (WCB) regions during 2001 to 2019 whereas the long-term changes are insignificant in the other regions of BoB. Significant increase in AOD in the NWB and WCB regions is mainly contributed by dominant outflow of anthropogenic emissions from Indo-Gangetic Plain (IGP) area of Indian subcontinent. The magnitude of AOD decreased by half from northern BoB to equatorial region due to increase in distance from the source region. The contribution of anthropogenic AOD was >70% to total AOD with higher contribution during winter and lower during summer. The rate of increase in both total and anthropogenic AOD were close to 0.104 and 0.099 per decade in the NWB and 0.069 and 0.059 per decade in the WCB region between 2001 and 2019. The rate of increase in total and anthropogenic AOD decreased from 2001 to 2009 (0.164 and 0.115 per decade respectively) to 2010-2019 (0.068 and 0.076 per decade respectively) in the NWB region. Significant increase in anthropogenic AOD by 50 and 30% was observed during El Nino and La Nina periods respectively than normal year in both NWB and WCB regions due to change in strength and direction of winds. Though some fraction of anthropogenic AOD is found over the entire BoB, significant rate of increase in anthropogenic AOD is found only about 23% of the area of BoB than hitherto reported as entire BoB. The impact of atmospheric deposition of anthropogenic aerosols on biogeochemical processes, such as primary production and ocean acidification, needs further evaluation.

Key words: Aerosol optical depth, pollution, long-term trends, Indo-Gangetic Plain, Bay of Bengal

1. Introduction

The role of aerosols on global climate change is well recognised, however the factors influencing their variability is poorly understood (IPCC, 2001). Aerosol Optical Depth (AOD) is a measure of the extinction of the solar radiation by particles in the atmosphere (dust, smoke, pollution) due to either absorption or scattering of light. Satellite-based measurements of AOD provide valuable information of region of their source and rate of increase over period through continuous measurements. Since AOD indicates the absorption of sunlight induced by aerosols (haze), direct measurement of aerosol loading contributes in rating of global climate change (Zhao et al., 2008). Numerous studies were carried out to examine the regional and global variability in AOD using multiple satellite observations, which includes the Moderate Resolution Imaging Spectroradiometer (MODIS), and Multiangle Imaging Spectroradiometer (MISR) (Zhang and Reid, 2010; Hsu et al., 2012; Pozzer et al., 2015; Klingmüller et al., 2016; Mehta, 2015; De Leeuw et al., 2018; Proestakis et al., 2018; Che et al., 2019). Despite many studies on AOD, the controlling factors responsible for inter-seasonal and decadal variability of regional AOD is unclear (Zhao et al., 2008; Zhang and Reid, 2010).

The northern Indian Ocean is unique as it is covered three sides by Asian land mass where one third of the global population is dwelling close to the coast. This region is occupied by the fast-growing cities and therefore high contribution of anthropogenic aerosols are being produced (eg: Kumar et al., 2008; Moorthy et al., 2010; Nair et al., 2014). Seasonal variations in the position of the Inter-Tropical Convergence Zone (ITCZ), which locates south of equator during winter and north of 25°N during summer, is dependent on the cross-equatorial atmospheric energy transport, the equatorial net energy input (Keshtgar et al., 2020) and monsoon circulation (Schott and McCreary, 2001). The seasonal migration of the ITCZ allow to spread pollutants over the Indian Ocean (Ramanathan et al., 2001). In contrast, weak horizontal pressure gradients and winds prevail during the dry period (March-May and October), when the aerosol residence time is expected to be long due to absence of efficient aerosol removal mechanisms (mainly washout and rainout).

In the past few decades, emissions from various pollutants and their components are increasing in the fast growing and expanding countries (such as India and China) due to enhanced industrial activity and agricultural practices. Cohen et al. (2014) reported large temporal and spatial variations in AOD in the Southeast Asia, where one of the most complicated cloud-precipitation systems exists with complex meteorological conditions and well defined geographic features (Reid et al., 2013). Furthermore, frequent widespread biomass burning events occur in the Southeast Asia (Taylor et al., 2018) leading to emission of huge amounts of aerosols into the atmosphere. Increased population along with these factors make Southeast Asia as a hot spot for atmospheric pollution. Ramachandran et al.

(2005) reported high-atmospheric absorption of radiation over the BBoB due to presence of higher soot content. Zhang and Reid (2010) reported the highest rate of increase in AOD over the BoB (0.076 per decade), followed by coastal China Sea (0.062 per decade), and Arabian Sea (0.06 per decade) between 2000 and 2009. They further stated that appreciable share of human induced sources is contributed to the total AOD over the BoB and South China Sea than other regions in the globe. Numerical models suggested that marginal increase in anthropogenic AOD (by 0.04–0.08) is possible over India by 2030 (Das et al., 2020). Though higher rates of AOD is reported between 2000 and 2009 in the BoB, the rates may be modified during recent decade (2010-2019) from that of earlier. In addition, seasonal reversal in wind direction occurs in the BoB, between winter and summer monsoons, that may result in possible change in source and rate of increase in AOD over the BoB.

The spatial trends in AOD were studied under aegis of Amidst Integrated campaign for aerosols, gases, and radiation budget over India (ICARB) during March-May 2006 (Nair et al., 2008). Prior to ICARB, limited studies were conducted to estimate the concentration of aerosols over the BoB during February and October 2003 (Satheesh et al., 2006; Ganguly et al., 2005; Vinoj et al., 2004; Sumanth et al., 2004) and the constant observations from Port Blair, Andaman Islands, India (Moorthy and Babu, 2006). Apart from this, some additional measurements were also made in the southern BoB, close to Sri Lanka under the aegis of Indian Ocean Expedition (INDOEX) program (Nair et al., 2008).

and... Sinha et al. (2011) studied size distribution of the aerosol over the BoB and reported that the aerosols are mainly sub-micron size (0.25 to 0.3 μm) highlighting the strong anthropogenic influence in the northern and Indian coastal BoB whereas much higher sized (0.4 to 0.6 μm) particles are found over open oceanic regions. Dey and Singh (2002) evaluated seasonal distribution of size of the aerosol particle and reported that existence of unimodal nature during winter and bimodal nature during summer over the BoB. They further found that dominant finer fraction of aerosols over the BoB than Arabian Sea. Although some studies were made on AODs over the BoB (Satheesh et al., 2006; Vinoj et al., 2004; Sumanth et al., 2004) they were mainly concentrated over western BoB around the Indian subcontinent. The first ever study was the ICARB of 2006 to cover significant part of BoB (Nair et al., 2008a; 2009; Moorthy et al., 2009) however they did not cover the eastern part of BoB, which is more affected by anthropogenic emissions. The present study is an effort to fill these gaps with constant observations made over the entire BoB region encircled between 83°E and 97°E and 0°N and 23 °N for past 19 years. As per our knowledge, this would be a first attempt over the oceanic region around BoB featuring total, anthropogenic, and natural AOD along with their size distribution. The objective of this study is to 1) evaluate the long-term variability in total, natural and anthropogenic fraction of AOD for the past two decades (2001-2019) with reference to space over the BoB, 2) identify the hot-

spot region where the impact of anthropogenic aerosol deposition is more and 3) discuss the possible implication on biogeochemistry of the BoB due to aerosol deposition over the surface waters.

2. Methodology and Study area

Total aerosol optical depth (AOD) data with 1 x 1° grid resolution at 470 nm, 550 nm and 860 nm wavelengths are obtained from Moderate Resolution Imaging Spectroradiometer Terra satellite (MODIS) collection 6.1 and level 3 during the period of 2001 to 2019 (<https://ladsweb.modaps.eosdis.nasa.gov/search>). The Level 3 (L3) MODIS AOD atmosphere products data is a global-gridded science product. The fine mode fraction of AOD is computed from spectral AOD and fine-model AOD (given as aerosol_optical_Depth_small-Ocean) (Levy et al., 2013). The error on the total AOD is $\pm 0.03 + 5\%$ of AOD (Wei et al (2019)). The fraction of anthropogenic AOD from total AOD is estimated following Kaufman et.al. (2005). The estimation of anthropogenic component from MODIS total AOD uses several assumptions. They are a) the fine fraction of aerosol is constant to the AOD for a given aerosol type. Smoke and pollution is assumed to be mostly contributed to fine aerosols, whereas dust and marine components contribute to coarse aerosols. b) All smoke is considered to be anthropogenic origin and all dust is originated naturally. c) It is assumed that wild fires contributes ~20% of biomass burning and anthropogenic sources contribute up to 10% of dust. d) MODIS derived fine fraction is highly correlated with true fine fraction and errors are small. e) Based on AERONET and MODIS analysis (Kaufman et al., 2001, 2005), the baseline marine aerosol optical thickness is assumed to be 0.06 ± 0.01 under calm conditions.

According to Kaufman et al. (2005) total aerosol optical thickness (τ_{550}) is defined as sum of anthropogenic (τ_{anth}) from pollution and smoke + dust (τ_{dust}) + baseline marine (τ_{mar}) sources. The same can be written as

$$\tau_{f=550} = f_{550} \tau_{550} = f_{anth} \tau_{anth} + f_{dust} \tau_{dust} + f_{mar} \tau_{mar}$$

Here f_{mar} , f_{dust} , f_{anth} are fraction fine particles from marine, dust and anthropogenic sources and they are derived from MODIS whereas τ_{mar} is determined from AERONET. The total fine fraction (f_{550}) and optical thickness (τ_{550}) were derived by MODIS. Based on this,

$$\tau_{anth} = [(f_{550} - f_{dust}) \tau_{550} - (f_{mar} - f_{dust}) \tau_{mar}] / (f_{anth} - f_{dust})$$

The complete details of the estimations and constants used are given in Kaufman et al. (2005). In present study we used MODIS collection 6.1 AOD data whereas Kaufman et al. (2005) used collection 4 data to derive the constants, therefore we calculated the constants f_{mar} , f_{dust} , f_{anth} using MODIS collection 6.1 data for the same regions and time periods that of Kaufman et al. (2005). We obtained

f_{mar} , f_{dust} , f_{anth} are 0.32, 0.48, 0.77 respectively and the computation of fraction of anthropogenic aerosols carry an uncertainty of up to $\pm 30\%$. The natural AOD is estimated as difference between total AOD and anthropogenic AOD (Kaufman et al., 2005).

The Ångström exponent is computed following Knobelspiesse et al. (2004) using 470 and 860 nm bands data as given below,

$$\text{Ångström exponent (AE)} = - \ln (\text{AOD}_{470}/\text{AOD}_{860})/\ln (470/860).$$

ERA-Interim reanalysis of monthly wind vectors data with a resolution of $1 \times 1^\circ$ grid during the period between 2001 and 2019 is downloaded from <https://apps.ecmwf.int/datasets/data/interim-full-moda/levtype=sfc/>.

In order to derive long-term changes, monthly mean data of total, natural and anthropogenic

AOD is used for weighted linear regression analysis with variability in the selected regions given in Fig. 1. In order to evaluate decadal variability, the data collected between 2001 to 2009 and 2010 to 2019 were separated and the rate of increase for these two decades are evaluated separately and combined the entire data to obtained the mean trends for both decades (2001-2019).

3. Results and Discussion

3.1. Temporal and spatial variability in total AOD over the BoB

Earlier studies based on MODIS data (2005–2010) indicated a steady sharp increase over the western BoB in AOD values when compared with southern BoB (Spandana et al., 2012). Many studies have reported that aerosols loading over the BoB are high compared to Arabian Sea and South Indian Ocean (Nair et al., 2013; Ramachandran 2005; Dey et al., 2004).

Wind blow dominantly from northeast during winter (November, December, January and February), northwest during spring (March and April) and southwest during summer and fall monsoon seasons (May to October) over the BoB (Fig. 2). As a result, the western BoB receives aerosols from Indian subcontinent whereas eastern BoB receives from the southeast Asia during winter. On the other hand BoB receives pollutants from southeast of India during summer and fall monsoons. Nair et al. (2008) reported that the aerosol over the head BoB (mostly north western part) is sourced from the emissions coming out of biomass burning transported from Myanmar, whereas fossil fuel emissions from the east coast of India dominates the southern BoB. Spatial variations in total, anthropogenic and natural AOD showed well-defined meridional and zonal variability over the BoB with reference to wind direction (Fig. 3 -5). Perennial high total AOD is noticed over the head BoB (north of 20°N) and northwestern BoB (0.5 to 0.6) with higher levels during spring (March, and April) and the least during

fall monsoon period (October) (Fig. 3). Total AOD decreased steadily toward south with mean AOD of ~ 0.2 (Fig. 3). The total AOD progressively increased from September to June in the northwestern BoB with maximum during June. The spreading of total AOD from northwest to southeast is noticed mostly during all months. Higher total AOD is noticed over the central Bay during May to July and the least during November to January. Except in equatorial region, the maximum total AOD loading is observed during May-June (0.6-0.4), which is due to transportation of aerosols by summer monsoonal winds (Fig. 3). . Tiwari et al. (2016) reported high aerosol loading during June and low during November over entire BoB. Seasonal variability shows relatively lower AOD values during winter (November to February) due to stable meteorological conditions that slows down the long-range transportation of aerosol from settlement areas. The seasonal wind data suggests aerosols are brought from the north and northeastern India during winter whereas from southwest direction during summer (May to September) (Fig. 2). The highest mean seasonal aerosol loading was reported in the BoB during June–August with an enormous contribution of dust coming from Middle East, Arabia and southwest Asia (Priyith et al., 2013; Kaskaoutis et al., 2011; Satheesh et al., 2010; Dey and Di Girolamo, 2011). The aerosols from southeastern India and marine sources contribute during summer as dominant marine winds prevail during transition period (March, April and October). The decrease in total AOD during July to September is caused by monsoonal precipitation over BoB and also over the Indian subcontinent (Soman and Kumar, 1990) resulting in washout from the atmosphere (Fig 2). Moorthy and Babu (2006) reported that transmission of aerosols to the BoB varies significantly with the seasonal and synoptic-scale circulation patterns. Therefore, increased total AOD during November to May with peak in May is not contributed by the same source. Hence, the fraction of anthropogenic and natural to total AOD may be variable during different seasons due to change in direction of winds.

3.2. Spatial and temporal variations in anthropogenic and natural aerosols over the BoB

The spatial and temporal distribution of natural and anthropogenic AOD is contrasting to each other due to reversing in wind direction due to monsoon (Fig. 5 and 4). The anthropogenic AOD (Fig 4) increased rapidly from September to March with higher in the northwest and spread towards northeast by March. Unlike total AOD, the maximum in anthropogenic AOD is found during February and minimum during May-August (Fig. 4). This pattern together with wind direction suggests that dominant northwest winds during winter brought anthropogenic aerosols from the Indo-Gangetic Plain (IGP). The least anthropogenic aerosols are found over the BoB during summer (Fig. 1 and 2) due to heavy precipitation over the northeastern India (1000-2500 mm; Soman and Kumar, 1990). Since the major source of anthropogenic aerosols comes from Indo-Gangetic Plain, they have been washed-out from the atmosphere due to heavy precipitation resulting in decrease in anthropogenic contribution

over the BoB during summer (Fig. 4). In contrast, dominant distribution of natural AOD over anthropogenic sources were noticed during summer in the entire BoB and the least during winter (Fig. 4 and 5). The dominant natural AOD during summer is caused by removal of anthropogenic sources through rainfall to over the BoB (Fig. 5). In addition to this, the winds blow from southwest direction, where industrial activities are much lower compared to the IGP that resulted in low contribution of anthropogenic fraction. The high levels of total AOD noticed during May-July are mainly contributed by natural AOD (Fig. 2). This study suggested that dominant contribution from anthropogenic AOD is noticed during winter and fall monsoon especially in the west and northwestern BoB whereas dominant source from natural aerosols occurred during summer in the entire BoB.

3.4. Spatial and temporal variations in size of aerosols over the BoB

For the classification of types of aerosols on monthly basis over the study regions, Ångström coefficient value is computed and depicted in Figure 6. The higher values of coefficient (> 1) represent absorbing (anthropogenic/secondary/fine) type of aerosols sourced from dust and biomass burning whereas the lower values (<1) indicate the existence of non-absorbing (natural/primary/coarse) aerosol particles (Prospero et al., 2002). Higher coefficient (>1) was noticed in the entire northern BoB during October to March indicating anthropogenic aerosols were dominated during fall and winter and lower coefficient values (<1) were observed during May to September. This suggests dominant contribution of natural aerosols (coarser) during summer (Fig. 5) and presence of finer particles during winter. Remarkable longitudinal gradients over the BoB, with increase in fine aerosols towards east, indicating that anthropogenic aerosols are transporting over from Indian subcontinent to the eastern BoB (Fig. 6). This gradient was more noticeable in the northern than southern BoB suggesting transmission of aerosols from the west. Higher value of Aerosol Index associated with a lower value of small mode fraction aerosol particles over northern BoB was reported by Sreekanth et al. (2013). However, higher Ångström coefficient value was found during March represents the fine particles and that could be mainly due to the long range transport of aerosols mainly emitted from wheat crop residue burning over the IGP and it is consistent with the earlier reports (eg: Kaskaoutis et al., 2011; Nair et al., 2010).

3.5. Long-term monthly variability in different fractions of AOD

Based on spatial variability in total, anthropogenic and natural AOD, the BoB is divided into 5 regions based on the specific characteristics of the region with reference to source of AOD. They are 1) north western BoB (NWB; 18-23°N, 84-89°E), where anthropogenic aerosols are received from IGP, 2) Western Coastal BoB (WCB; 14-19°N, 83-88°E), where lower anthropogenic aerosols are received, compared NWB, due to less industrial activities along east coast of India, 3) Andaman Sea (AS; 10-

15°N, 92-97°E) region, where anthropogenic aerosols are received dominantly from Southeast Asia, 4) Central BoB (CB; 10-15°N, 85-90°E), where long-range transported aerosols are received from south and southeast Asia and 5) equatorial region (ER; 0-5°N, 85-90°E), where perennial low aerosols are received as this zone is away from the land mass, hence this zone is relatively pristine within BoB (Fig. 1). Despite the selected regions are unique with reference to variable source of aerosols, but they showed significant intra-annual variability. Hence the long-term trends are examined at monthly time-scale in the selected region in the BoB.

Table 1 shows the rate of monthly mean total, anthropogenic and natural AOD change in the selected regions over BoB using the 19- years of MODIS aerosol data. The weighted linear regression analysis with variability in data was applied to the 19-year of MODIS using monthly mean AOD values in each region.

3.5.1. *Northwestern BoB (NWB)*

The climatological mean levels of total, anthropogenic and natural AOD are depicted with reference to month in Figure 7a in the NWB region. The total AOD increased from September to May and decreased during summer (June to August) with maximum during May.. The contribution of natural to total AOD increased from 3 to 65% from April to August and decreased linearly to 4% from September to March. On the other hand, higher contribution of anthropogenic to total AOD is noticed (62 to 86%) between September and April and decreased to 35 to 52% during summer (Fig. 7a). The magnitude of variability in anthropogenic AOD, between the lowest in July (0.2) and the highest in March (0.5), was high (0.3) in the NWB. Kumar et al. (2008) reported that dominant contribution of anthropogenic aerosols during winter and spring monsoon over in the northern BoB compared to the Arabian Sea. The significant enhancement in the AOD over the IGP and the adjoining regions is due to the pollutants, smoke and biomass burning associated with dust advected from the arid western Asian regions which in turn influencing the NWB region (Beegum et al. 2008). Moreover, as the transported mineral and anthropogenic aerosols seems to have a dominant particle size of $\sim >1 \mu\text{m}$, it would have contributed toward the finer particles and thereby resulting in the higher values of AODs (Fig.7a).

Significant increase in rate of total AOD was observed between 2001 and 2019 (0.160 to 0.164 per decade) during March to April and minimum during July and August (0.029 to 0.034 per decade) (Table. 1). The rate of increase in total AOD was significant during January to April and October to December at 99% confidence level whereas it was insignificant (<90% confidence level) during summer (May to September) in the NWB region (Table. 1). In the NWB region, where the highest

values of AODs were recorded, a wide spread of atmospheric particles over the Indian subcontinent, covering all of central India and the IGP, are providing channels for dispersion of continental aerosols, mostly of mixed type (continental and marine). The IGP area, which is considered as hub of emissions (anthropogenic) from coal- based power plants, industries, transport, mining, urban and agricultural activities, account for ~21% of the land area of the Indian subcontinent (eg: Nair et al., 2007; Kumar et al., 2008). Several studies have also reported the dominance of high AOD over this region during winter (Prasad et al., 2006; Jethva et al., 2005; Singh et al., 2004; Ramana et al., 2004). The large loading of aerosols in this region is due to prevailing westerly winds combined with the orography of the IGP that spatially restricts the aerosols into a rather narrow pathway leading to the discharge into the northern and head BoB (Nair et al., 2007; Niranjana et al., 2007; Girolamo et al., 2004). Significant increase in anthropogenic AOD values were found throughout the year with the high during January to June and December (0.104 to 0.376 per decade) and low during July to November (0.02-0.090 per decade) (Table 1). The decrease in the anthropogenic AOD during summer is caused by change in the direction of winds from north west to southwest and also monsoonal precipitation resulting in washout from the atmosphere (Fig. 2). The low anthropogenic AOD during summer in the NWB is contributed by long-range transport as well (Das et al., 2012, Gogoi et al., 2008). The rate of increase in anthropogenic aerosols are significant during entire year at variable confidence level (Table 1).

Decrease in natural AOD is observed during all months in the NWB region (-0.009 to -0.12 per decade), however these rates are statistically insignificant, except during September and October (Table 1). This study suggests that anthropogenic AOD contributed significantly to the total AOD throughout the year and natural AOD did not show significant variability indicating that significant increase in aerosols over the NWB region is contributed by anthropogenic sources. The mean rate of increase in AOD is >10 times higher than that in the equatorial BoB (Table 1).

3.5.2. *Western Coastal BoB (WCB)*

The climatological mean levels of total, anthropogenic and natural AOD were displayed similar monthly distribution pattern from that of NWB with higher rate of increase in total AOD during January to April and October to December (0.058 to 0.143 per decade) and statistically significant at >90% confidence level. In contrast lower rate of increase in total AOD was observed during May to September (0.010 to 0.053 per decade; Fig. 7b) and they are statistically insignificant (Table 1). The contribution of natural to total AOD varied between 4 and 78% with higher during May to August (70-73%) and lower during October to February (4-14%; Fig. 7b). Higher contribution of anthropogenic to total AOD was noticed (22 to 82%) between September and April (57-82%) associating with higher Ångstrom coefficient (>1; Fig. 6) and it was decreased to 22 to 30% during summer. The magnitude

of variability in anthropogenic AOD from the lowest in July to the highest in March was 0.25 in the WCB. The significant rate of increase in anthropogenic AOD is found during February to September (0.031 to 0.179 per decade) and it is insignificant during other months. The long-term variability in natural AOD is statistically insignificant during all months in the WCB, except during September. The low rate of increase in both total and anthropogenic AOD in the WCB than NWB is caused by wind direction as most of the aerosols are transported to the open sea region compared to the coastal zone. The WCB region adjoins the highly polluted eastern coastal India with three major harbors, such as Visakhapatnam, Paradip and Haldia (Figure 1) and several industrial activities along the coastal Bay. As a result, high anthropogenic aerosol levels are prevailed over WCB regions due to local activities, in addition to the pollutants enters from the IGP. The observed moderately high values of anthropogenic and total AOD is the consequence of these two factors.

3.5.3. *Andaman Sea (AS)*

The level AOD in the AS region is lower than NWB by 1.7 times but the temporal variability is almost uniform compared to the other selected regions (Fig. 7c). The contribution of anthropogenic AOD varied between 16 and 88% with higher during October to April (71-88%) than May to September (17-37%) and contrasting to that is noticed in case of natural AOD (9-29% and 61-83% respectively). The magnitude of variability in anthropogenic AOD from the lowest in July to the highest in March was 0.21 in the AS. Based on wind pattern, the AS region receives atmospheric aerosols from the continental regions of East Asia and south China (Fig. 2). Previous reports from the BoB exhibits the role of advection of air parcels from South China and East Asia and in the increment of the AOD over Port Blair, Andaman Islands (Vinoj et al., 2009; Nair et al., 2009; Moorthy and Babu, 2006; Moorthy et al., 2003). Though increasing trend in both total and anthropogenic AOD is noticed but they are not statistically significant, except in April (Table 1). Streets et al. (2003) have reported increase in urban activities as the reason for enhancement in anthropogenic emissions over East Asia. The higher Ångstrom coefficient in the AS region during winter further supports significant amounts of anthropogenic sources to this region (Fig. 6).

3.5.4. *Central BoB (CB)*

The CB region received relatively low magnitude of aerosols from the continent compared to NWB and WCB but close to that of AS. The monthly variability suggests that higher total AOD during May to August is associated with natural AOD whereas anthropogenic AOD increased between October and April (Fig 7d). The contribution of anthropogenic to total AOD varied from 63 to 84% during October to April whereas 12 to 35% during other months. The magnitude of variability in

anthropogenic AOD from the lowest in July to the highest in March was 0.17 in the CB. In contrast, natural AOD contributed between 65 and 87% during May to September and lower (15-32%) during other months. Figure 2 suggests the air parcels crossing CB were found to have originated from the western coastal regions of the East Asia, which might transport human induced aerosols along with the natural marine aerosols, because these air parcels have travelled across the vast oceanic regions. The moderate values of total and anthropogenic AOD and fairly low values of natural AOD throughout the year might be due to mixed type of aerosols (sea salt spray and anthropogenic aerosols). The total and anthropogenic AOD found in the CB region is lower than 2 and 2.5 times respectively than NWB region (Fig. 7d). The CB region showed significant increase in total and anthropogenic AOD by 0.43 to 0.088 and 0.049 to 0.103 per decade respectively during January to April whereas the long-term trend was statistically insignificant during other periods.

3.5.5. Equatorial Region (ER)

The least AOD levels were noticed in the ER region compared to other regions in the BoB due to away from the terrestrial sources leading to dilution of natural and anthropogenic aerosols (Fig. 7e). Despite very low levels of AOD the monthly pattern of all components of AOD remained same as that of other regions in the BoB. Even in the ER region, the contribution of anthropogenic AOD is higher during October to April (37-63%) than other months (10-23%) (Fig. 7e). The total and anthropogenic AOD are 2.7 and 5.6 times lower in the ER compared to NWB region respectively. The magnitude of variability in anthropogenic AOD from the lowest in July to the highest in March was 0.10 is noticed in the ER. Since winds crossing ER were mostly from oceanic region, they would be mostly dominated by a larger amount of coarse mode sea salt as marine aerosols associated with sea spray and smaller amounts of fine aerosols. (Fig 6). Anthropogenic aerosol concentrations were low in southern BoB as it opens to the vast Indian Ocean. The rate of change in total, anthropogenic and natural sources are statistically insignificant in the ER region.

3.6. Long-term variability in total, natural and anthropogenic AOD and recent modifications

Zhang and Reid (2010) reported an increase in anthropogenic and total AOD over entire BoB as 0.058/decade and 0.07/decade between 2000 and 2009 respectively whereas the present study suggests that trend differs with space and time (Table 1). Relatively higher levels (2-4 times) of total and anthropogenic AOD were observed in the NWB and WCB regions compared to ER region where the lowest levels were noticed (Fig 7). The rate of increase in both total and anthropogenic AOD is 6 to 10 times higher in the NWB and NCB regions compared to other regions in the BoB due to proximity to the source in the former regions and direction of winds. Despite ER region is far from the source

region, still significant fraction of the total AOD is contributed by the anthropogenic AOD suggesting that entire BoB is contaminated with anthropogenic AOD. However, the rate of change in neither natural nor anthropogenic AOD is significant in CB, AS and ER regions suggesting that significant modification in atmospheric composition is unlikely in these regions. Though NWB and NCB regions are highly influenced by total and anthropogenic AOD, the rate of increase is decreased from earlier decade (2001-2009) to recent decade (2010-2019) (Table 2). For instance, the weighted linear regress analysis suggested that the rate of increase in total and anthropogenic AOD in the NWB region was 0.164 and 0.115 per decade respectively during 2001-2009 and it is decreased to 0.068 and 0.076 per decade in the recent decade (2010-2019) respectively. Similarly the total and anthropogenic AOD in WCB was increased at the rate of 0.128 and 0.085 per decade respectively during 2001-2009 and it is decreased to 0.038 and 0.049 per decade during 2010-2019 respectively (Table 2; Fig. 8). Significant decrease in total AOD in both NWB and WCB are contributed by decrease in natural AOD. On the other hand, the rate of increase in total and anthropogenic AOD in the AS was 0.044 and 0.034 per decade respectively during 2001 to 2009 but the rate is insignificant during 2010-2019. The mean rate of increase in total and anthropogenic AOD during past two decades (2001-2019) was higher (0.104 and 0.099 per decade respectively) in the NWB than earlier report for the entire BoB for the period of 2001-2009 (0.07/decade and 0.058/decade respectively; Zhang and Reid, 2010) but same as that of in the WCB (0.069 and 0.059 per decade respectively; Table 2). This study suggesting that significant increase in total and anthropogenic AOD is found only in the NWB and WCB regions and further noticed that the rate of increase is decreasing in the recent decades. The area significantly contaminated with atmospheric pollutants in the BoB is about 23% ($0.47 \times 10^{12} \text{ m}^2$) of the entire Bay of Bengal ($2.02 \times 10^{12} \text{ m}^2$) than hitherto hypothesized as entire Bay.

3.7. Influence El Nino on total and anthropogenic AOD in the BoB

El Nino and La Nina modifies significantly the direction and strength of winds due to variations in the atmospheric pressure. The climatological monthly anomaly winds during El Nino and La Nina were presented over the northern Indian Ocean in Figure 9. Reversing of wind direction is noticed during all seasons during El Nino and La Nina period over the BoB. For instance, winds blow from north eastern direction during January and March during El Nino period and the same was changed to southwest during La Nina period (Fig. 9). Similarly east or south easterly winds are noticed during August and November in El Nino period whereas it was westerly and north westerly during La Nina period (Fig. 9). The change in direction of winds during El Nino and La Nina modifies the source and magnitude of pollutants received over the BoB. Out of 19 years considered in this study (2001-2019), 8 years were recorded as El Nino (2002, 2004, 2006, 2009, 2014, 2015, 2018 and 2019) and La Nina

(2001, 2005, 2007, 2008, 2010, 2011, 2016 and 2017) and 3 were as normal years (2003, 2012 and 2013). Figure 10 depicts the mean total, anthropogenic and natural AOD in the NWB and WCB regions during El Nino, La Nina and normal years. It was observed that about 50% increase in anthropogenic AOD is noticed during El Nino period from that of normal year in both NWB and WCB regions whereas it was increase by ~30% during La Nina compared with normal years (Fig. 10). The contrasting to that was noticed for natural AOD, which is increased from El Nino followed by La Nina and normal years. This pattern could be explained with the change in wind directions as dominant north westerly or north easterly winds were noticed during El Nino whereas south westerly during La Nina period suggesting that dominant terrestrial (anthropogenic) during former and marine source during latter period compared to normal period (Figs. 2 and 9) (Li et al., 2011). Therefore extreme events enhance pollutants over the western BoB than normal periods.

3.8. Implications of aerosols on ocean biogeochemistry

In recent years, dramatic economic rise, rapid industrialization, increase in population growth, traffic flow, construction and demolition projects along the coastal areas critically affected the atmospheric environment, especially with regard to aerosols loading (UNDP report 2018). Due to close proximity of these areas with coastline of BoB, it is directly as well indirectly affecting the coastal ecosystem along with atmosphere. Many previous studies have reported that this increase in aerosols loading over BoB region is due to excessive and uncontrollable emissions from IGP region of India and from neighbouring southeast Asian countries (Babu et al., 2013). These aerosols are impacting the biogeochemistry of BoB waters in due course of time. Sarma et al. (2015) reported that artificial enrichment of western coastal BoB through deposition of sulphates and nitrates led to rapid acidification by 3 to 4 times from that of open ocean region. Significant alterations in plankton community due to acidification is already reported in the coastal BoB (Biswas et al 2017; Biswas et al., 2011). Biswas et al. (2017), based on microcosm experiments, reported abrupt increase in the diatom and tintinnids (Biswas et al., 2012) as a result of increasing CO₂ in the BoB coastal waters.

Enhanced pollutants in the atmosphere decreases the temperature in the lower atmosphere and also sea surface temperature (SST) due to reflecting the incoming radiation (Ramanathan et al., 2001; 2007; Sarma et al., 2015). Dong et al. (2014) reported that significant cooling of atmospheric temperature and SST due to anthropogenic aerosols over the BoB using CMIP5 models. They found that competing forcing roles of warming by green-house gases is compensated by cooling by anthropogenic aerosols. Sarma et al. (2015) reported decrease in -0.7°C of SST during winter between 1991 and 2011 is reported and attributed to increase in atmospheric pollution. The modifications in SST and air

temperature would modify sea-to-air exchange of heat and mixing that would ultimately impact the nutrients availability in the mixed layer.

Significant increase in primary production (3-33% in the coastal region; Yadav et al., 2016 and 13% in the open ocean BoB; Srinivas et al., 2011a) is reported due to deposition of atmospheric nutrients. In addition, alteration of phytoplankton community structure is noticed due to deposition of nutrients from the atmosphere leading to modifications in N:P stoichiometric ratios (Yadav et al., 2016). This would modify prey-predator relationship and food web dynamics. Though this study suggests that significant increase in atmospheric aerosols over the NWB and WCB regions, however, the modifications in the chemical composition with time is unknown due to lack of data. Since significant increase in anthropogenic AOD in the NWB and WCB is noticed since past two decades, it is possible that dissolved inorganic and organic nitrogen deposition on the surface waters may be enhanced resulting in elevated N:P ratios that may modify phytoplankton composition and primary production. In order to predict possible changes in ecosystem due to atmospheric aerosols, long term observations in elemental composition of atmospheric aerosols are highly essential in the BoB.

Increase in SST anomalies are reported during El Nino period due to warming of ocean resulting in weakening of mixing (Murtugudde et al., 2000; Sarma, 2006) in the Indian Ocean. This may lead to decrease in nutrients inputs to the surface leading to decrease in Chl-a as evidenced from the negative Chl-a anomalies during El Nino period (Murtugudde et al., 2000). Sarma (2006) estimated that ~30% decrease in primary production in the Arabian Sea during 1997-98 El Nino compared to normal period due to weakening of mixing and nutrients input. In contrast, our study suggests that enhanced atmospheric aerosols may possibly increase anthropogenic nutrients input to the surface ocean that may compensate decrease in nutrients supply through vertical mixing during El Nino. The ground truth observations with composition of atmospheric aerosols are needed during both El Nino and normal period in order to confirm the hypothesis. Nevertheless this study suggested that anthropogenic aerosols are increasing significantly in the NWB and WCB regions whereas significant modification in the ecosystem structure is possible and further studies are required to confirm these changes..

Summary and Conclusions

Increase in atmospheric pollution over Bay of Bengal (BoB) and South China Sea is reported between 2001 and 2009 due to increase in industrial activity in the South and Southeast Asia. However, long-term variability in spatial and temporal variability in atmospheric pollutants over the BoB is unclear. Spatial and temporal variability in total, natural and anthropogenic aerosol optical depth (AOD) over the BoB is examined between 2001 and 2019. Significant increase in both total and anthropogenic

AOD is noticed in the northwest (NWB) and western coastal BoB (WCB) and it is statistically insignificant in the other regions of BoB due to dominant outflow of anthropogenic emissions from Indo-Gangetic Plain (IGP) area in the former regions. About 50-85% of the total AOD is contributed by anthropogenic fraction and it is higher during winter and lower during summer. The rate of increase in both total and anthropogenic AOD were 0.104 and ~0.10 per decade respectively in the NWB and 0.069 and 0.059 per decade respectively in the WCB region between 2001 and 2019 with relatively higher during 2001-2009 than 2010-2019. It was further noticed that increase in 50% (30%) of anthropogenic AOD during El Nino (La Nina) period than that of normal year is noticed in both NWB and WCB regions due to change in strength and direction of winds. Our study suggests that significant impact of anthropogenic AOD is noticed only in the 23% of area (NWB and WCB) of the BoB than hitherto reported as entire BoB. The impact of deposition of anthropogenic elements, such as dissolved organic and inorganic nitrogen, phosphorus and sulphates on ocean acidification and primary production need to be evaluated in future in the NWB and WCB regions.

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Declarations

Ethics approval and consent to participate

Not Applicable

Consent for publication

Not Applicable

Availability of data and materials

We used remote sensing sensors data and the web links where data are available are given

Competing interests

Authors do not have any competing interests

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Authors' contribution

KY – processing of total AOD data, preparation of figures and initial manuscript preparation

VDR – computation of natural, and anthropogenic contribution of AOD, preparation of figures and discussion during manuscript preparation

SB- Processing of AOD data, preparing trends, and involved in manuscript preparation

VVSS – concept of the work, guidance and finalizing the manuscript.

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Legends to Figures

Figure 1: The study region showing major ports along the Bay of Bengal coast, major polluted regions/sources, the regions (NWB, WCB, CB, AS and ER) are also depicted.

Figure 2: Spatial and monthly distribution of climatological mean of wind direction and speed in the BoB

Figure 3: Spatial and monthly distribution of climatological mean of total AOD in the Bay of Bengal

Figure 4: Spatial and monthly distribution of climatological mean of anthropogenic AOD in the BoB

Figure 5: Spatial and monthly distribution of climatological mean of natural AOD in the BoB

Figure 6: Spatial and monthly distribution of climatological mean of Ångstrom exponent in the BoB

Figure: 7: Monthly mean variation in total, anthropogenic and natural AOD in the selected location in the BoB. The standard deviation of the data is given as vertical bar.

Figure 8: Long-term trends in total, anthropogenic and natural AOD in the NWB and WCB regions in the BoB. The standard deviation of the data is given as vertical bar.

Figure 9: Spatial and monthly distribution of wind anomalies during (a) El Nino and (b) La Nina periods in the BoB

Figure 10: Mean levels of total, anthropogenic and natural AOD during El Nino, La Nina and normal years in the NWB and WCB regions in the BoB. The standard deviation of the data is given as vertical bar.

Figure 1:

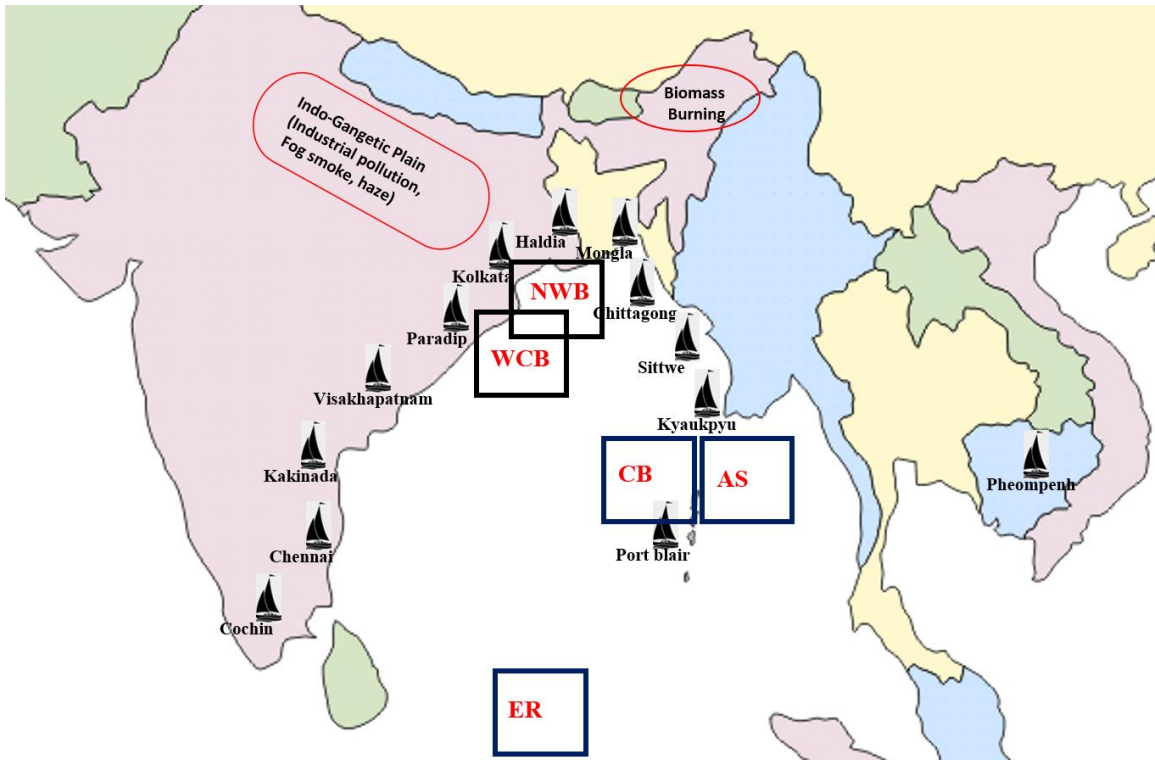


Figure 2:

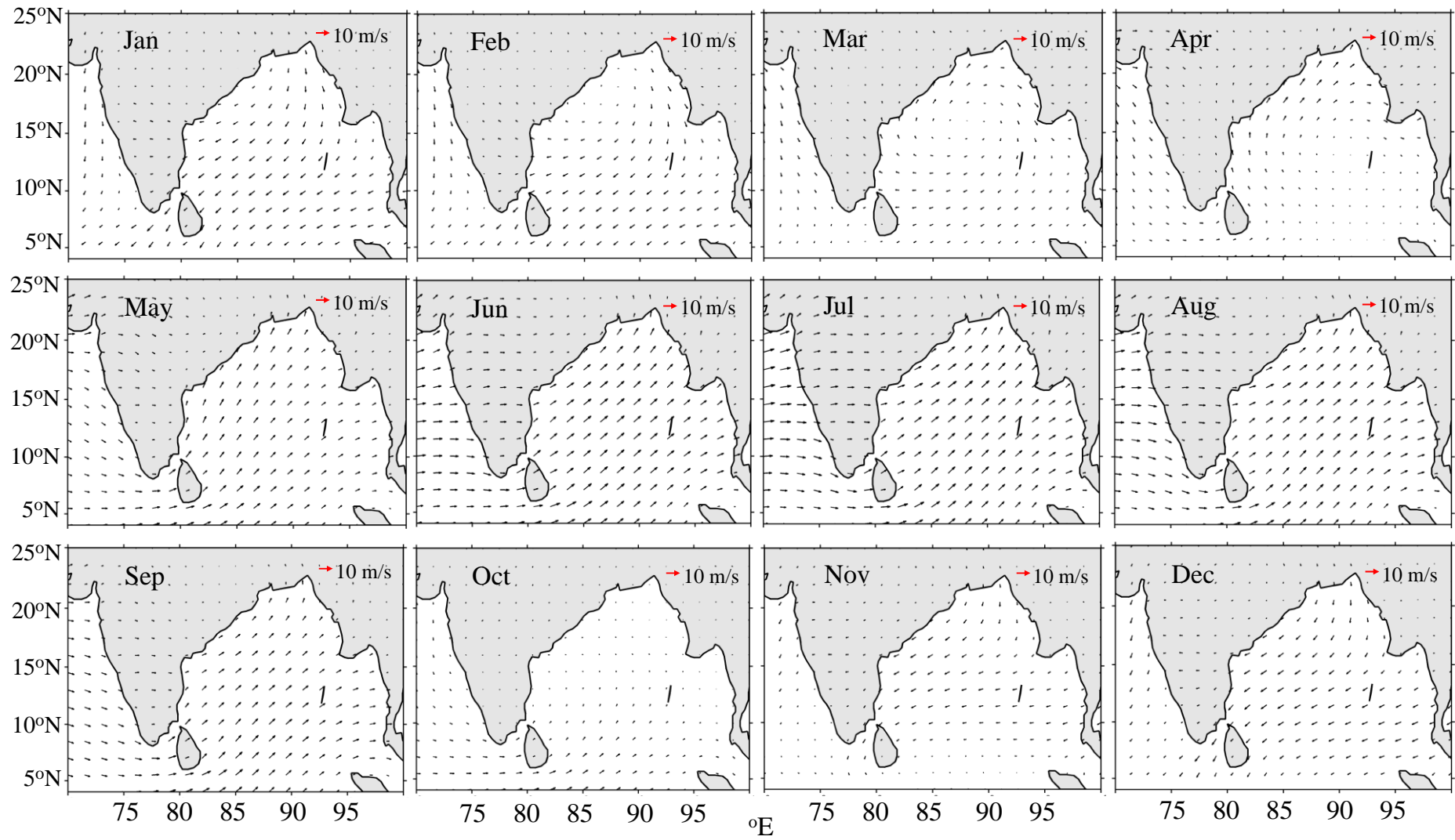


Figure 3:

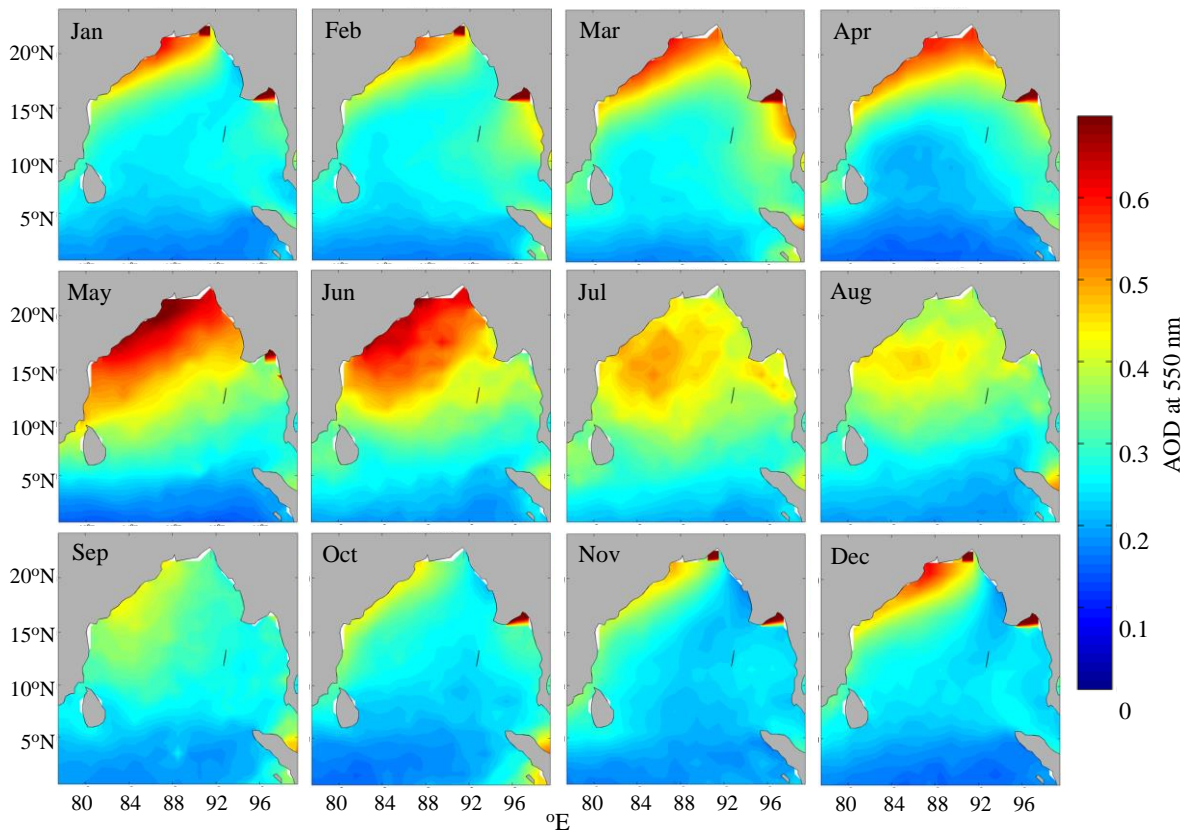


Figure 4:

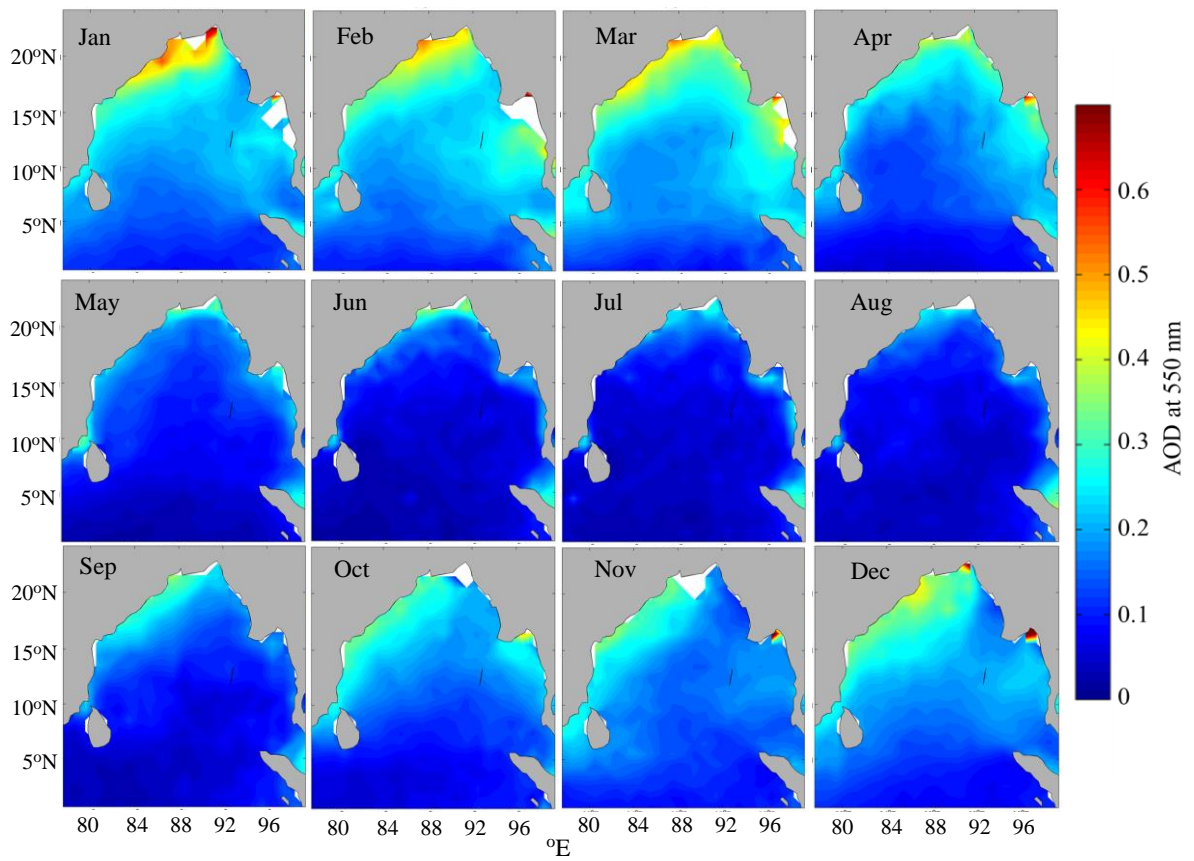


Figure 5:

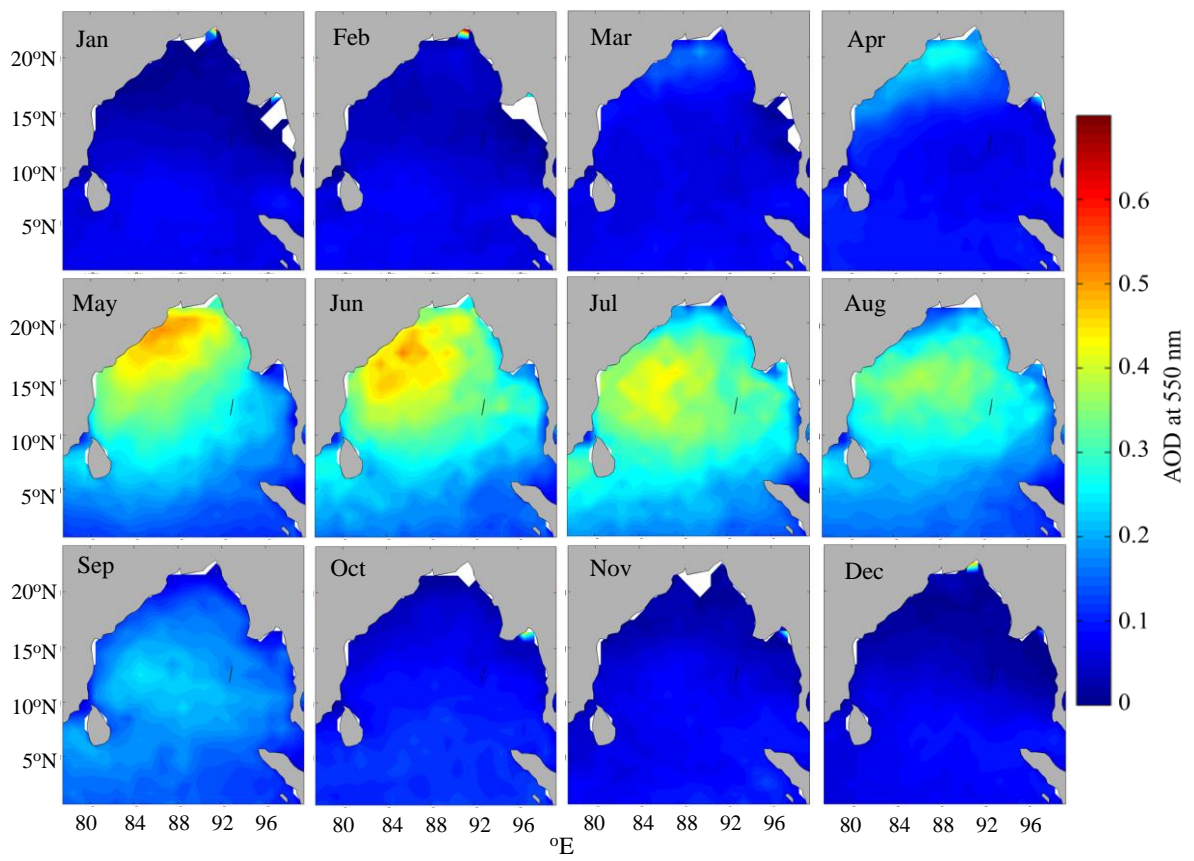


Figure 6:

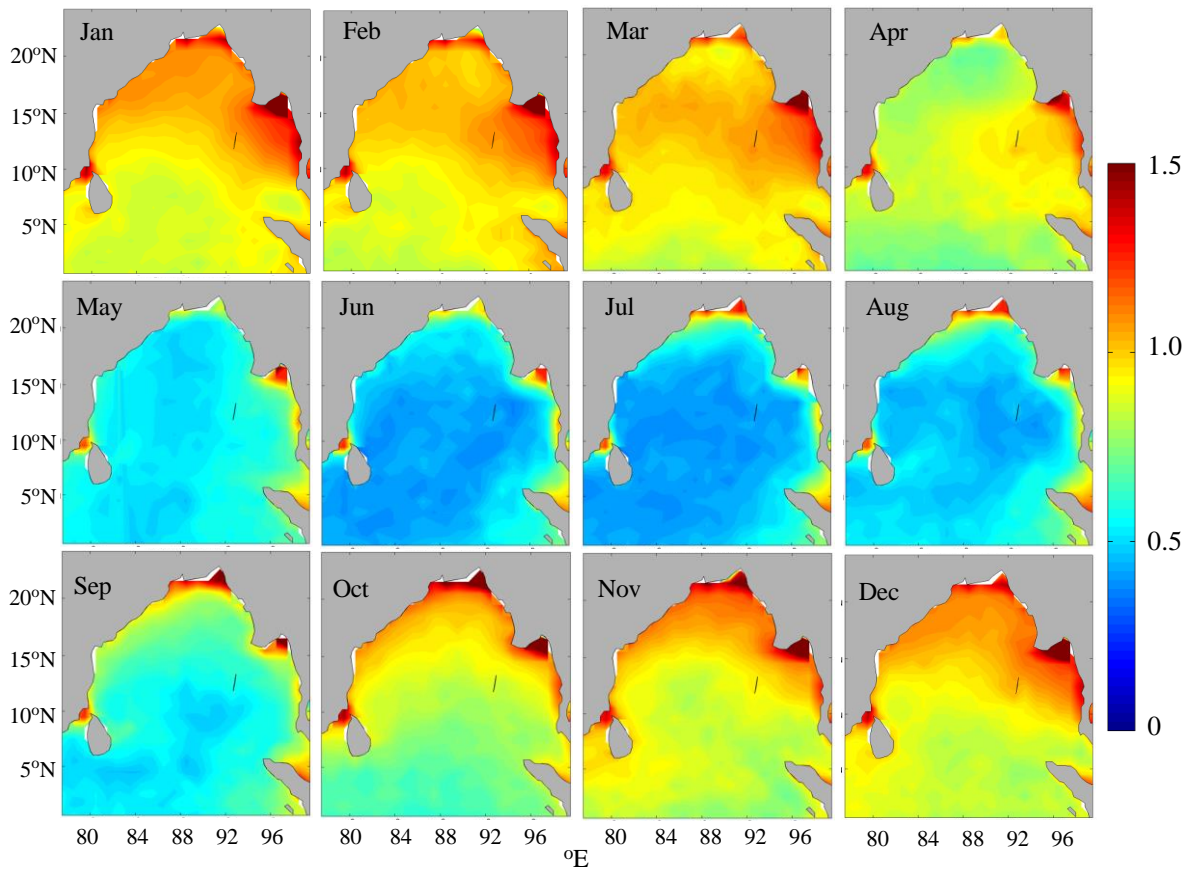


Figure: 7:

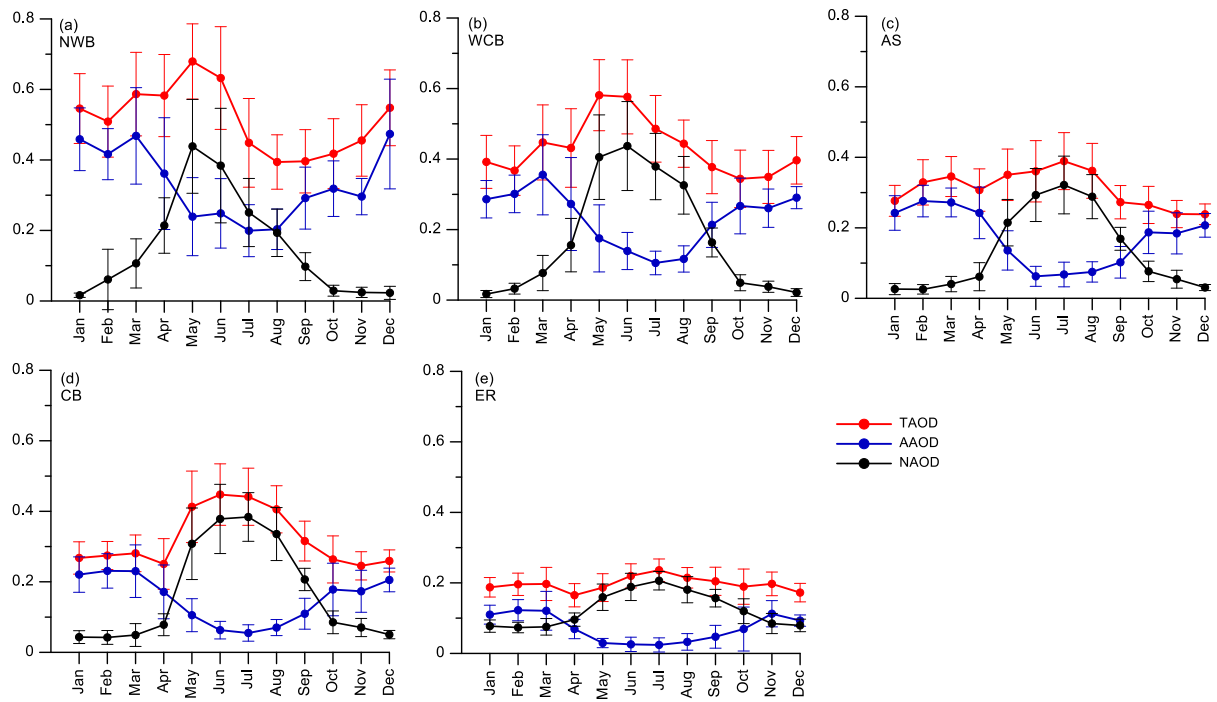


Figure 8:

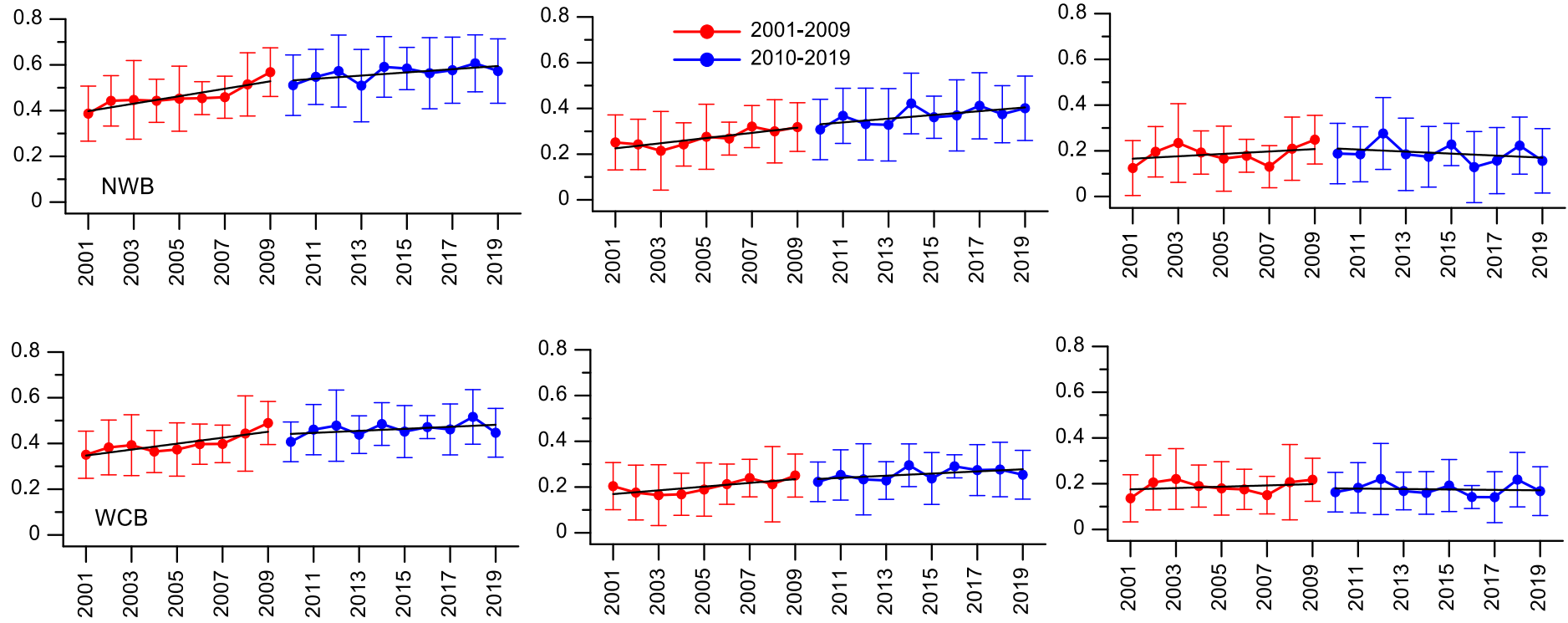


Figure 9a:



Figure 9b:

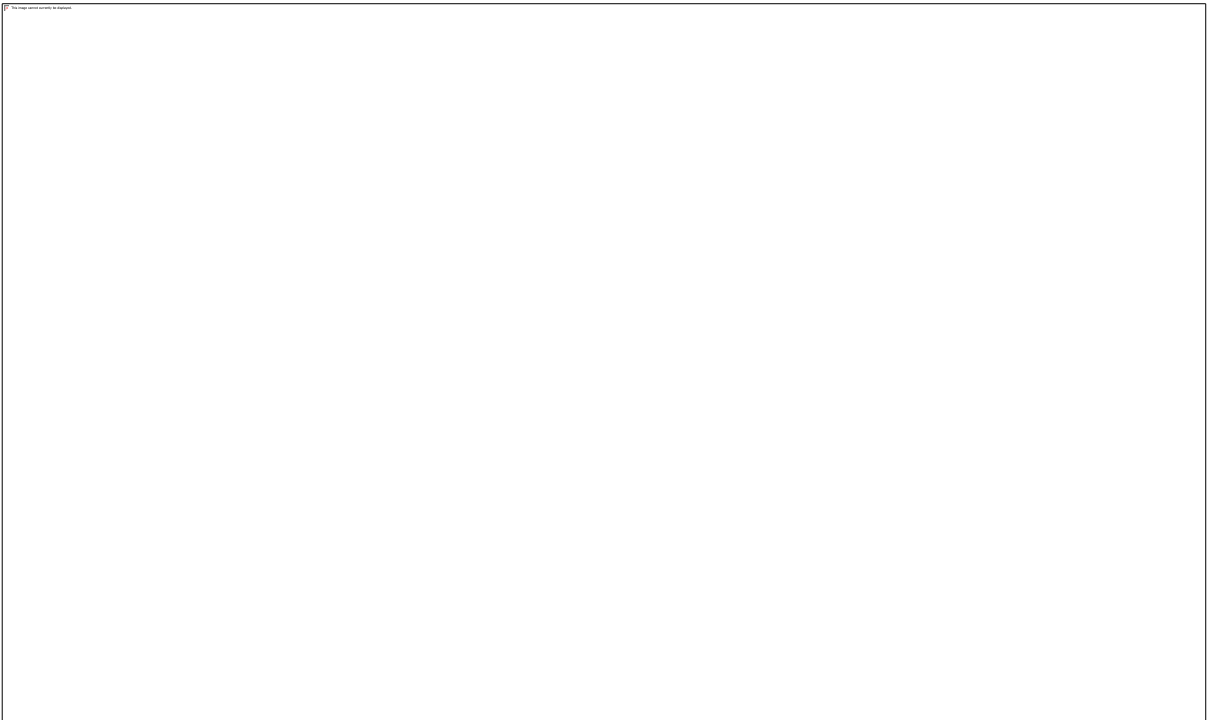


Figure 10:

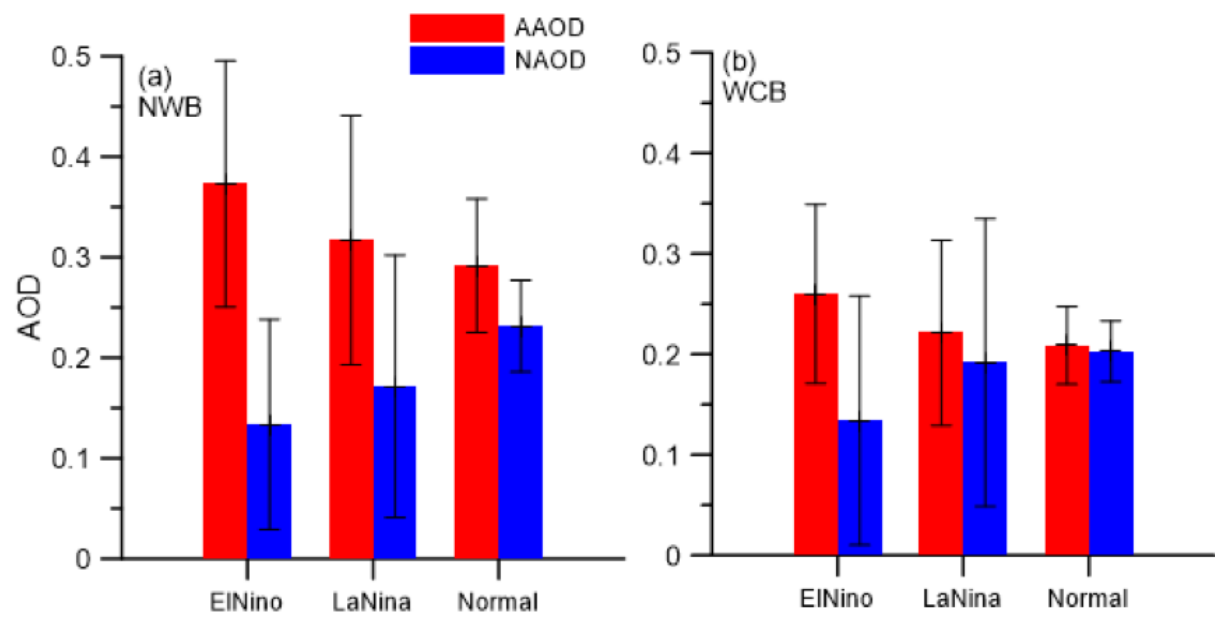


Table 1: Rate of monthly mean variations in total, anthropogenic and natural AOD data for last 19 years (2001-2019) in the selected regions in the BoB.

Region	Month	Total AOD Per decade	P values	Anthropogenic AOD Per decade	P values	Natural AOD Per decade	P values
18-23N 84-89E(NWB)	Jan	0.131	0.001	0.321	0.01	-0.028	NS
	Feb	0.148	0.001	0.104	0.02	-0.009	NS
	Mar	0.164	0.001	0.152	0.001	-0.028	NS
	Apr	0.160	0.001	0.222	0.001	-0.060	NS
	May	0.068	NS	0.158	0.001	-0.090	NS
	Jun	0.041	NS	0.131	0.001	-0.120	NS
	Jul	-0.034	NS	0.075	0.01	-0.079	NS
	Aug	0.029	NS	0.063	0.01	-0.017	NS
	Sep	0.066	NS	0.090	0.02	-0.046	0.01
	Oct	0.114	0.001	0.062	0.005	-0.019	0.01
	Nov	0.139	0.001	0.020	NS	-0.028	NS
	Dec	0.123	0.001	0.376	0.001	-0.049	NS
14-19N 83-88E(WCB)	Jan	0.099	0.001	0.014	NS	0.000	NS
	Feb	0.103	0.001	0.061	0.01	-0.011	NS
	Mar	0.120	0.02	0.117	0.01	-0.048	NS
	Apr	0.143	0.01	0.179	0.001	-0.029	NS
	May	0.015	NS	0.125	0.001	-0.099	NS
	Jun	0.034	NS	0.064	0.01	-0.033	NS
	Jul	0.053	NS	0.032	0.05	0.005	NS
	Aug	0.024	NS	0.031	0.02	-0.007	NS
	Sep	0.010	NS	0.054	0.02	-0.047	0.001
	Oct	0.058	0.02	0.048	NS	-0.012	NS
	Nov	0.085	0.01	0.022	NS	-0.002	NS
	Dec	0.079	0.001	0.024	NS	-0.002	NS
10-15N 92-97E(AS)	Jan	0.008	NS	0.012	NS	-0.001	NS
	Feb	0.009	NS	0.012	NS	0.000	NS
	Mar	0.035	NS	0.022	NS	-0.006	NS
	Apr	0.081	0.001	0.085	0.01	-0.012	NS
	May	-0.024	NS	0.048	NS	-0.069	0.01
	Jun	-0.017	NS	0.006	NS	-0.033	NS
	Jul	-0.003	NS	0.009	NS	0.011	NS
	Aug	0.045	NS	0.025	NS	0.021	NS
	Sep	0.033	NS	0.052	0.05	-0.024	NS
	Oct	-0.010	NS	-0.015	NS	0.001	NS
	Nov	-0.010	NS	-0.015	NS	0.018	NS
	Dec	-0.003	NS	0.004	NS	0.000	NS
10-15N	Jan	0.057	0.001	0.060	0.001	-0.014	NS

86-90E (CB)							
	Feb	0.043	0.001	0.049	0.01	-0.012	NS
	Mar	0.051	0.05	0.073	0.01	-0.024	0.02
	Apr	0.088	0.001	0.103	0.001	-0.005	NS
	May	-0.055	NS	0.041	0.05	-0.089	NS
	Jun	0.016	NS	0.011	NS	-0.004	NS
	Jul	0.000	NS	0.001	NS	0.014	NS
	Aug	0.007	NS	0.002	NS	0.006	NS
	Sep	0.021	NS	0.046	0.05	-0.029	0.02
	Oct	0.032	NS	0.046	NS	-0.009	NS
	Nov	0.034	NS	0.036	NS	-0.009	NS
	Dec	0.030	NS	0.021	NS	-0.007	0.05
0-5N 86-90E (ER)	Jan	0.024	0.05	0.020	NS	0.008	NS
	Feb	-0.001	NS	-0.002	NS	-0.003	NS
	Mar	-0.015	NS	-0.003	NS	-0.015	NS
	Apr	0.016	NS	0.010	NS	0.008	NS
	May	-0.020	NS	-0.006	NS	-0.006	NS
	Jun	0.023	NS	-0.006	NS	0.015	NS
	Jul	0.020	NS	0.005	NS	0.012	NS
	Aug	0.016	NS	-0.019	NS	0.028	NS
	Sep	0.032	NS	0.036	0.02	0.003	NS
	Oct	0.061	NS	0.082	NS	-0.014	NS
	Nov	0.033	0.02	0.031	NS	-0.004	NS
	Dec	0.007	NS	0.003	NS	0.005	NS

Table 2: Annual mean variations rate of change in total, anthropogenic and natural AOD data for last 19 years (2001-2019) in the selected regions in the BoB based on weighted linear regress analysis.

Aerosols Fraction	18-23N 84-89E(NWB)		14-19N 83-88E(WCB)		10-15N 92-97E(AS)		10-15N 86-90E(CB)		0-5N 86-90E (ER)	
	Slope (/decade)	P value	Slope	P value	Slope	P value	Slope	P value	Slope	P value
2001-2009										
Total AOD	0.164	0.001	0.128	0.001	0.044	0.001	0.071	0.001	0.0183	NS
Anthropogenic AOD	0.115	0.001	0.085	0.001	0.034	0.001	0.037	0.01	-0.002	NS
Natural AOD	0.042	NS	0.025	NS	0.011	NS	0.034	NS	0.016	NS
2010-2019										
Total AOD	0.068	0.01	0.038	0.05	0.0002	NS	0.006	NS	-0.006	NS
Anthropogenic AOD	0.076	0.01	0.049	0.02	0.022	NS	0.037	0.05	0.014	NS
Natural AOD	-0.040	NS	-0.005	NS	-0.009	NS	-0.024	NS	-0.014	NS
2001-2019										
Total AOD	0.104	0.001	0.069	0.001	0.015	NS	0.029	0.05	0.016	NS
Anthropogenic AOD	0.099	0.001	0.059	0.001	0.015	NS	0.038	0.001	0.010	NS
Natural AOD	0.002	NS	-0.007	NS	-0.005	NS	-0.011	NS	0.006	NS