Atmospheric Iron and Aluminium Deposition and Sea-Surface Dissolved Iron and Aluminium Concentrations in the South China Sea off Malaysia Borneo (Sarawak Waters)

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ABSTRACT

South China Sea (SCS) is an oligotrophic sea which usually receives low nutrients supply. However, massive atmospheric dust input was occurred during the haze event in Southeast Asia for almost every year. The input of dissolved iron (DFe) and dissolved aluminium (DAl) from dust and nearby land into SCS off Sarawak Borneo region during the worst haze event in 2015 of the Southeast Asia were investigated. The estimation dust deposition during this study was 0.162 mg/m^2/yr. The atmospheric fluxes of total Fe and total Al at the offshore Sarawak waters were 0.611 µmol/m^2/yr and 2.03 µmol/m^2/yr, respectively, where the readily available dissolved Fe and Al from the dust were 0.11 µmol/m2/yr (DFe) and 0.31 µmol/m^2/yr (DAl). Fe has higher solubility (17.78%) than Al (15.21%). The lateral fluxes (e.g. from the nearby land) were 37.08 nmol/m^2/yr (DFe) and 125 nmol/m^2/yr (DAl), with strong Fe organic ligand class L_1 (log K:22.43 – 24.33). High concentrations of DFe and DAl at the surface water of the offshore region, coincided with high concentration of macronutrients due to the prevailing south-westerly winds originated from the west Kalimantan. Low residence times, ~0.92 (DFe) and ~1.31 (DAl) years, corresponded well with DAl$_{excess}$ in surface seawater due to biological utilization of DFe. Future works emphasize on natural organic Fe(III) ligands and phytoplankton study are needed for better understanding on biogeochemistry of Fe and Al at SCS off Malaysia Borneo.

Keywords: Atmospheric dust input, haze event 2015, Sarawak EEZ waters, South China Sea, trace metals

INTRODUCTION

Vegetation and peatland wildfires occurred every year across the Equatorial Asia, particularly at the Southeast Asia, during the dry season between September and October. The 2015 wildfire that started in Indonesia Borneo and then spread to Sarawak (Malaysia Borneo) began in July, resulted in thick smokes that blanketed Southeast Asia countries, such as, Malaysia, Brunei, Indonesia, Thailand and Singapore. It was likely the worst haze event since 1997 (Koplitz *et al.,* 2016). The combustion activities were worsening by drought conditions during the El Niño and the Indian Ocean Dipole, where the fire emissions can be rose up to 30 times greater than during La Niña (Crippa *et al.,* 2016). Moreover, the prevailing winds transported the wildfire smokes for hundreds to thousands miles away from the originating fires on the land to the sea, thus, the spatial and temporal effect of atmospheric inputs on surface seawater biogeochemistry may be seen even in remote oceanic areas.

South China Sea (SCS) is the largest marginal sea with oligotrophic condition (Wen *et al.,* 2006) which receives low nutrients supply (Guo *et al.,* 2012), where the nutrients may come from different sources such as coastal and atmospheric inputs (Chen *et al.,* 2021). Studies showed that there were significant inputs of micronutrients (*i.e.* Fe and Al) from atmospheric dust deposition in SCS due to the strategic location of SCS (Lin *et al.,* 2007; Wong *et al.,*

2002; Wong *et al.*, 2007; Du *et al.*, 2021). However, Fe data from Sarawak waters off the SCS were absent since approximately 20 years ago, with the available data was from 1996 and 1997 (Boonyapiwat, 2000), while Al data was absent since then.

Fe is very important element for marine organisms, which Fe acts as a key element for biological processes to take place including photosynthesis, uptake of nitrates, fixation of N_2, and detoxification of reactive oxygen species in seawater (Berman-Frank *et al.*, 2004; Rijkenberg *et al.*, 2008; Shi *et al.*, 2020). Pico-phytoplankton is the most tolerate taxa to low nutrient condition in oligotrophic seawater. However, even this taxa is also affected when the supply of Fe and other nutrients are very low. Fe in seawater is originated from various sources such as atmospheric (*e.g.* aerosol) dust, riverine and sediment inputs. Meanwhile, Al is known as an ideal tracer to indicate the dust input into the surface seawater because it is a major and invariant component of continental materials and also it has low solubility with a range of 1.5 to 10 % in rainwater and seawater, respectively (Gehlen *et al.*, 2003). In addition, strong evidence showed that Al was driven primarily by dust deposition as surface Al concentration was directly proportional to dust deposition (Measures & Vink, 2000). Other significant inputs of Al into the seawater is riverine run-off while sediment input is minimum since scavenged Al from surface water is biologically recycled (Kraemer, 2004).

Therefore, the objective of this study was to investigate the input of Fe and Al from the atmospheric dust deposition and from the nearby land into the Sarawak waters off the SCS during the worst haze event in 2015. The research approach is to use the trace metal concentrations to estimate the solubility, residence time, atmospheric and lateral metal fluxes at the study region.

MATERIALS AND METHODS

Study Sites and Samples Collection

Samples collection were done in Sarawak EEZ waters on board of *MV SEAFDEC2* between 17th of August and 5th of October 2015. A total of 12 stations were selected for surface seawater sampling, where stations were divided into three

areas (*i.e.* Area I (nearshore), Area II (mid-shore) and Area III (offshore)) (Figure 1). For aerosol samples, four samples were collected around Areas I and II while five samples were collected around Area III. For the aerosol samples, PM_{10} were collected from the bridge of the ship within 6 hours (Zhang *et al.*, 2007) when the ship moved (to avoid contamination from the exhaust smoke) by using a low volume pump air sampler (MiniVol portable air sampler) equipped with 0.45 μm of membrane filter (Sartorius Polyamid, 37070) and calibrated to a flow rate of 10 L/min using bubble flow meter. The filter papers were placed in clean double zipper plastic bags and then in acid-washed container labelled with sampling details. Filter papers for Fe and Al analyses were suspended into 25 mL of ammonium acetate buffer solution (pH 4.7) for 60-120 minutes to release the aerosol samples. Then, the supernatants were filtered using 0.2 μm cellulose acetate filter papers and acidified with 0.08 M HNO_3 in 50 mL vials for dissolved Fe (DFe) and dissolved Al (DAl) analyses, meanwhile the unfiltered supernatants were used to analyse total concentrations of Fe (TFe) and Al (TAl). Fe and Al concentrations were determined by using inductively coupled plasma optical emission spectrometry (ICP-OES) (Perkin Elmer, Optima 8000). Filter papers that were not exposed to the dust were used as blanks and treated the same way as samples (Sarthou *et al.*, 2003).

Surface seawater samples were collected using Van Dorn water sampler (5.0 L) at ~3 m depth. All 500 mL samples were filtered (0.45 μm) and stored in acid-washed bottles, then acidified to pH ~2 using 12 M HCl (R&M chemical analytical reagents) and were kept in double re-sealable zipper plastic bags. For Fe speciation analysis, 250 mL of filtered seawaters were collected into high density polyethylene bottles (Nalgene) and immediately frozen at -20 °C (not acidified) for subsequent land-based analysis.
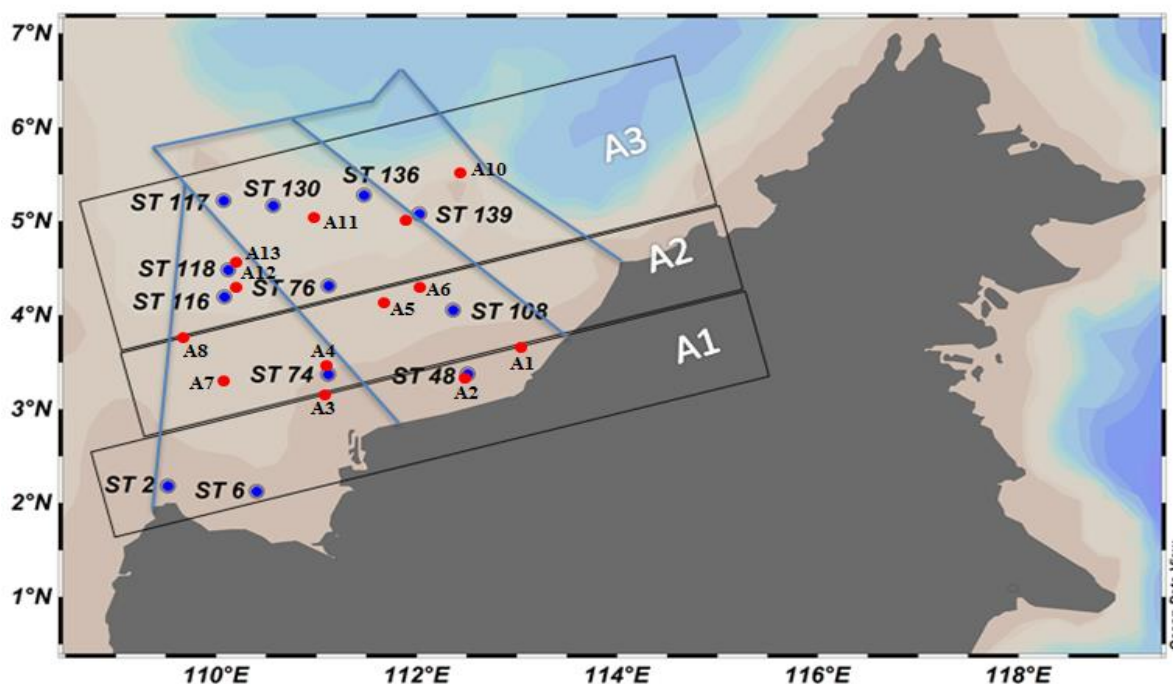
Trace Metal Analysis

Analysis of DFe conducted following the method provided by Idrus (2016), while for DAl, analysis conducted was followed the method by Brown and Bruland (2008). Off-line pre-concentration technique was applied using commercially available chelating resin with

IDA-type functional groups Toyopearl AF Chelate 650M resin (Part No. 19800, Tosoh Bioscience). Prior to pre-concentration, samples pH was adjusted to 4.5 (Fe) and 5.75 (Al) by adding 0.4 M ammonium acetate buffer and analysed using ICP-OES (Perkin Elmer, Optima 8000) (Ali and Shakrani, 2014). Certified Reference Material (CASS-5) was used in the Fe calibration procedure. The percentage recovery obtained for Fe was 95 %. For Al, percentage recovery was determined by using the internal

standards. The internal standard solutions were prepared using the treated assigned water samples in the same way of samples. The percentage recovery for Al obtained was 90 %.

The Fe speciation analysis was undertaken using competitive ligand exchange-adsorptive cathodic stripping voltammetry (CLE-ACSV) with TAC (thiazolylazo-p-cresol; Sigma-Aldrich) as competing ligand (Mohamed et al., 2011).

Figure 1. Map of sampling locations located at Sarawak water during *MV SEAFDEC 2* research from 17 August to 5 October 2015. A1: Area I; A2: Area II; A3: Area III. Blue dots: Surface waters sampling stations; Red dots: Aerosol sampling stations; Blue line represents the area of Sarawak waters

Estimation of solubility, residence time, atmospheric and lateral metal fluxes

Solubility of Fe in aerosol samples was measured as according to Chance *et al.* (2015). Assuming that the study areas were in steady state conditions, the atmospheric dust deposition (D) in the study area can be estimated by using DAl data obtained from this study following the method as explained in de Jong *et al.* (2007). Then, the atmospheric dust deposition flux was used for calculating of total trace metal atmospheric flux ($F_{atm, TM}$). Dissolved metals ($F_{atm, DTM}$) fluxes were calculated using obtained value of total flux of trace metals. Residence time is the average length of time that trace metals or any elements retained in the water column before removed from the water

column (in the steady state condition). Residence times of trace metals were calculated by using the average of DFe and DAl at the surface water multiplying with the mixed layer depth and then divided by the F_{atm},D_{TM}.

The horizontal fluxes of trace metal (F_{DTM}) were calculated by using the distance from the station located very close to the shore and the scale (the distance takes for the inshore concentration to drop by 1/e was estimated from the ln ([Fe]) or ln ([Al])) (Aguilar-Islas *et al.,* 2013) (Fe, 285.0 km; Al, 158.0 km). The exponential fit of $[DFe] = 4.048e^{x/285.0}$ while $[DAl] = 2.0728e^{x/158}$. Then, the parameterization was used to estimate lateral diffusion coefficient (K_h, m^2/s).
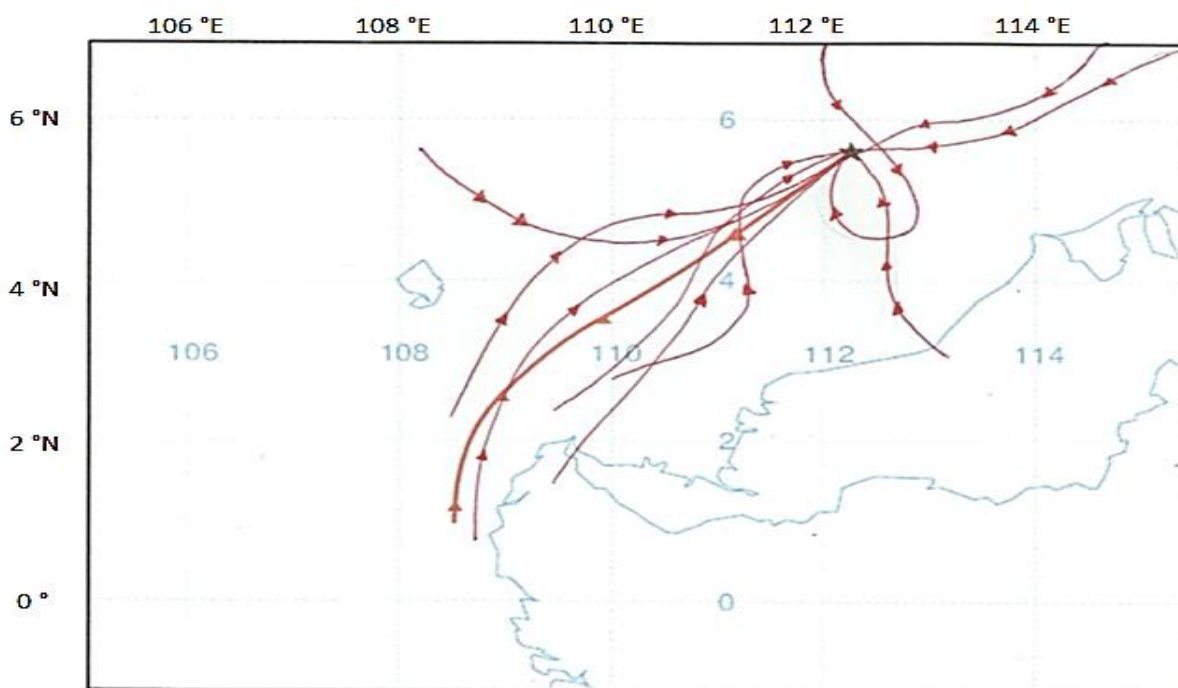
Other Calculation

Pearson's correlation coefficient (r) and scatterplots were used to quantify linear relationships between variables. Correlation coefficients were compared to the Pearson critical value for significance level p = 0.05 to test whether observed correlations were significant.

RESULTS AND DISCUSSION

The back trajectory analysis was conducted to determine the origin of air masses and established the source-receptor relationships. The information on back trajectories were comprehensively described in National Oceanic and Atmospheric Administration (NOAA) web portal (http:// ready. arl. noaa. gov/ HYSPLIT. php). In this study, back trajectories were plotted by using free online available software of NOAA HYSPLIT model, by setting one point at Area III (the farthest station) to determine the origin of wind that move towards or pass through the study area (Figure 2). Backward trajectory confirmed that prevailing south-westerly winds were originated from west Kalimantan and passed throughout our study locations. Wind that originated from West Kalimantan would possibly bring together all the particles that produce from biomass burning activities at that area.

Figure 2. Backward trajectories of prevailing winds during sampling period. Black star is a point that set at Area III to determine the origin of wind that move towards or pass through the study area. Red arrow represents the direction of wind from origin towards the sampling area

Many efforts had been done to suppress fires, but they were more challenging during El Niño conditions and on peatlands (Cattau *et al.,* 2016). Smoke from biomass burning activities contain high concentration of biologically important macro- and micro-nutrients (*i.e.* trace metals) (Hapsari *et al.,* 2021; Ponette-Gonzalez *et al.,* 2016). Emitted gasses and particles would be delivered to downwind, in this case from West Kalimantan towards Sarawak water and deposited into the water column via wet or dry deposition. In addition, one volcanic eruption was reported on the 30th of August 2015 at Lakon-Empung, Sulawesi Indonesia. The ash plumes from this eruption rose to an altitude up to 3 km and transported to nearby area including Malaysian waters (Global Volcanism Programme, 2013).

DFe and DAl distributions in surface water are presented in Table 1. The DFe concentrations were ranged from 10 to 278 nM (*n*=12) while DAl concentration were ranged between 7.82 and 24.98 nM (*n* = 12). Overall, offshore stations were detected with high concentration of DFe and DAl, coincided with

high concentrations of Nitrogen (N) and Phosphorus (P) (*i.e.* Station 116 and Station 76) (Idrus *et al.*, 2017). Results showed that distribution patterns of DFe and DAl were: Area III > Area II > Area I, indicated that concentration of DFe and DAl were increasing relative to distance from shoreline and possibly due to input from aerosol deposition. Lin *et al.* (2009) and de Jong *et al.* (2007) suggested the area that further away from coastline has a greater impact of nutrient input from atmospheric aerosols because the chances of nutrient input from another sources are lower. Atmospheric aerosols have been hypothesized as an important nutrients source to stimulate biogeochemical activities in the SCS (Ho *et al.*, 2010; Jickells *et al.*, 2005; Lin *et al.*, 2009;

Wong *et al.*, 2002; Wong *et al.*, 2007; Wen *et al.*, 2006; Du *et al.*, 2021). The high DFe and DAl concentrations were coinciding with the maximum concentration of Chl-*a* (Idrus *et al.*, 2017) in Area I. However, the N and P at these stations were low (Idrus *et al.*, 2017). DFe concentration in seawater obtained in this study fall between concentration range reported by Utoomprurkporn and Snidvongs (1999) during Matahari Expedition in Sabah, Sarawak and Brunei waters. DAl concentrations obtained in this study were lower than the values in the Yellow Sea (Li *et al.*, 2013) and in the SCS around the Pulau Perhentian, Terengganu (Mohamaed *et al.*, 2019), as their study areas were affected by eddies that generated input from bottom sediment.

Table 1. The coordinates, dissolved Fe (DFe) and dissolved Al (DAl) concentrations for each station from 17 August 2015 to 5 October 2015 at Sarawak waters (surface water)

Area	Station	Longitude (°E)	Latitude (°N)	DFe (nM)	DAl (nM)
I	48	112.511	3.374	79.00 ± 0.34	11.08 ± 0.01
	6	110.122	4.480	47.00 ± 0.02	7.82 ± 0.03
	2	110.087	4.194	59.00 ± 0.20	7.82 ± 0.06
II	108	112.364	4.052	10.00 ± 0.34	13.86 ± 0.01
	74	111.227	4.575	62.00 ± 0.14	7.82 ± 0.01
III	139	112.027	5.075	81.00 ± 0.14	18.94 ± 0.03
	136	111.475	5.275	88.00 ± 0.08	18.94 ± 0.03
	76	111.122	3.371	278.00 ± 0.32	24.98 ± 0.01
	130	111.166	4.015	189.00 ± 0.21	16.79 ± 0.01
	117	111.126	4.311	105.00 ± 0.56	16.79 ± 0.11
	118	110.573	5.168	68.00 ± 0.36	15.46 ± 0.01
	116	110.078	5.219	224.00 ± 0.97	18.94 ± 0.03

Total Fe (TFe), total Al (TAl), DFe, DAl distribution in aerosol samples are shown in Table 2. High concentrations of TFe (6.65 ± 0.99 mg/m^3), DFe (0.96 ± 0.06 mg/m^3), TAl (1.41 ± 0.02 mg/m^3) and DAl (0.19 ± 0.01 mg/m^3) in aerosol samples were recorded at Station 9 (Area III). Similar pattern of Fe and Al distributions were observed between surface water samples and aerosol samples. Fe and Al concentrations in aerosol were higher at Area III, suggested that both elements were coming from similar origin during this study which could have related to the starting of Northeast monsoon event as suggested by Huang & Lin (2015) and Lin *et al.* (2007). This high deposition of dust brings together other nutrients into surface water which could enrich the productivity. Based on aerosol optical depth (AOD) observation around SCS, high AOD was found at offshore areas of China, Indochina, Sumatra and Borneo (Huang and Lin,

2015). Thus, dust deposition was dominated at offshore areas (Areas III and II) compared to near-shore area (Area I). Previous studies indicated that smokes from biomass burning in Borneo and Sumatra were the main sources of trace metals and nutrients content in aerosol at the southern SCS from August to October correspond to high aerosol optical thickness (AOT) with fine dust particles (Holloway *et al.*, 2002; Lin *et al.*, 2007).

The direction of wind was reversed (from Asia towards Peru) in El Nino period that cause course dust particles blown away from sea surface and increase the fine dust particles produced from biomass burning. Atmospheric dust input was recognised as an important source of trace elements including Fe and Al (Baker & Jickells, 2017; Strzelec *et al.*, 2020). Surface water nutrient may also derive from the

sediment via water mixing. Based on vertical distribution of DFe at study area (Idrus *et al.,* 2016), it showed that DFe concentration was higher at deeper water than surface water at Area I indicating that nutrient was significantly affected by input from sediment. Meanwhile, Areas II and III showed the opposite trend in which higher DFe were observed at surface water then decreased towards bottom indicated that these areas were less affected by input from

Table 2. The coordinates, TFe, DFe, TAl and DAl concentrations for each station from 17 August 2015 to 5 October 2015 at Sarawak water (aerosol)

Area	St.	Longitude (°E)	Latitude (°N)	TFe (mg/m^3)	DFe (mg/m^3)	TAl (mg/m^3)	DAl (mg/m^3)
I	1	113.022	004.020	5.15 ± 0.65	0.73 ± 0.01	0.41 ± 0.04	0.05 ± 0.01
	2	112.327	003.374	4.18 ± 0.34	0.58 ± 0.01	0.42 ± 0.09	0.06 ± 0.01
	3	111.127	003.131	4.27 ± 0.23	0.57 ± 0.03	0.42 ± 0.01	0.06 ± 0.01
	4	111.122	003.371	4.79 ± 0.81	0.68 ± 0.03	0.38 ± 0.12	0.05 ± 0.01
II	5	111.423	004.171	5.19 ± 0.21	0.72 ± 0.07	0.43 ± 0.09	0.06 ± 0.02
	6	111.203	004.396	4.81 ± 0.88	0.68 ± 0.01	0.45 ± 0.01	0.07 ± 0.01
	7	110.092	003.201	2.51 ± 0.09	0.35 ± 0.01	0.97 ± 0.07	0.15 ± 0.01
	8	109.599	004.171	3.26 ± 0.11	0.45 ± 0.01	1.00 ± 0.09	0.16 ± 0.01
III	9	112.027	005.076	6.65 ± 0.99	0.96 ± 0.06	1.41 ± 0.02	0.19 ± 0.01
	10	112.203	005.390	6.78 ± 1.11	0.94 ± 0.06	0.94 ± 0.06	0.15 ± 0.01
	11	111.029	005.027	8.99 ± 0.32	0.92 ± 0.05	0.87 ± 0.03	0.11 ± 0.01
	12	110.276	004.275	4.30 ± 0.13	0.60 ± 0.01	0.75 ± 0.09	0.12 ± 0.01
	13	110.123	004.480	4.25 ± 0.94	0.60 ± 0.02	1.05 ± 0.09	0.15 ± 0.02

[TFe] Total Fe; [DFe] Dissolved Fe; [TAl] Total Al; [DAl] Dissolved Al

sediment. Aerosol loading into surface water may increase the productivity by increasing the biological production (Mahowald *et al.,* 2018), as shown at the most oligotrophic area in SCS (*i.e.* the southern-central SCS) in which the upwelling activities were absent but the Chl-*a* were elevated in July-August, consistent with the increment of the AOT (Lin *et al.,* 2009). However, the amplitude of Chl-*a* was low. High Fe and Al concentrations at Area I may relate to nutrients sources from the surrounding land and from bottom sediment (Idrus *et al.,* 2016), as nutrients from bottom sediments were supplied throughout the water column during water mixing aiding by effect of strong wind of NE monsoon (Adiana *et al.,* 2014). In contrast, high concentrations of Fe and Al in Area III can be related to input from long-range transported atmospheric dusts. TFe, DFe, TAl, and DAl concentrations in this study were higher as compared to study by Hsu *et al.* (2014) at northern SCS, but considerably lower than value from the East Asian region (Hsu *et al.,* 2013). Distance from source of biomass burning possibly affects the distribution of trace metals in the aerosol samples. Guo *et al.* (2012) stated that impact of atmospheric input to increase the total biomass of phytoplankton was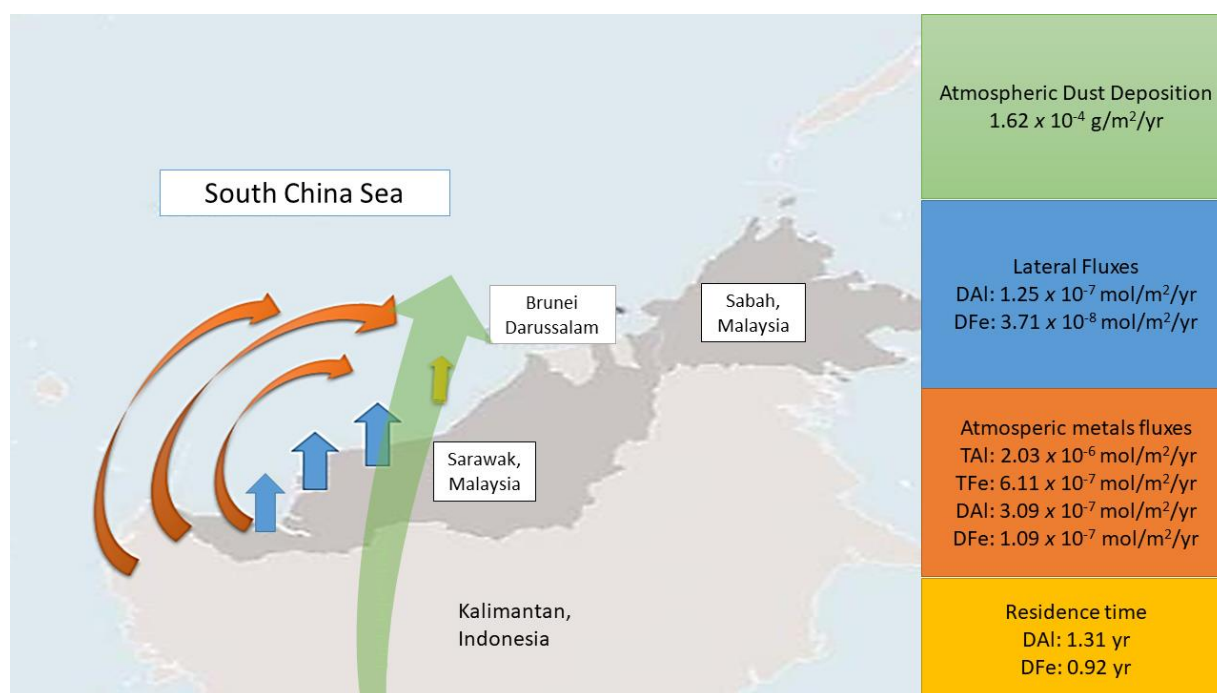 more significant in an oligotrophic open seawater rather than nutrient rich coastal region. These factors may be related to our study as there were absent of upwelling activities along Sarawak EEZ waters.

Solubility of Fe in aerosol at study location showed that Fe (17.78%) has a higher solubility than Al (15.21%), consistent with the study by Chance *et al.* (2015) which obtained Fe, Mn and Al solubility in aerosol (< 20%). This result may be related to input of anthropogenic materials sources from biomass burning activities. Atmospheric deposition into seawater occurs via two path either dry or/and wet deposition. In this study, atmospheric deposition was focused on dry deposition due to absent of rain water samples. In addition, the samplings were conducted during dry season thus dry deposition can be assumed as the main route of atmospheric deposition into the seawater. The atmospheric dust deposition calculated in this study is 1.62×10^{-4} g/m^2/yr. The total atmospheric flux of Fe and Al are 6.11×10^{-7} mol/m^2/year (Fe) and 2.03×10^{-6} mol/m^2/year (Al), while the dissolved atmospheric fluxes obtained in this study are 1.09×10^{-9} mol/m^2/year (Fe) and 3.09×10^{-7} mol/m^2/year (Al). Residence times obtained in this study are 0.92 years (DFe) and 1.31 years (DAl).

The relationship between concentration distributions of DFe and DAl relative to distance from shoreline showed that the concentration of both trace metals were increased exponentially with increasing distance from shoreline (R^2 = 0.8027, [DFe] (nM) = 33.899 $e^{0.002x}$) and (R^2 = 0.957, [DAl] (nM) = 5.6421 $e^{0.0017x}$). The horizontal fluxes were calculated to determine the rate of DFe and DAl fluxes from nearby land towards offshore. In this study, lateral flux of

DFe is 0.10 nmol/m^2/day or 37.08 nmol/m^2/year, and the lateral flux of DAl calculated is 0.35 nmol/m^2/day or 125 nmol/m^2/year. Comparison between DFe and DAl atmospheric fluxes with lateral fluxes showed that atmospheric input was the most prominent sources of DFe and DAl rather than lateral fluxes. Therefore, Fe and Al fluxes (*i.e.* from the atmospheric deposition and the lateral deposition) estimated in this can be summarized as in Figure 3 Fe that is

Figure 3. Inputs of Fe and Al into the SCS off Sarawak Waters during the haze event. Green arrow indicates the atmospheric dust deposition, orange arrows represent atmospheric metals (Fe and Al) fluxes, blue arrows show the latera fluxes from the land, and the yellow arrow is for the residence time of metals.

biologically available to be used by marine microorganism is in dissolved form. DFe distribution in surface water at our study area was significantly affected by atmospheric input indicated by the same patterns of DFe with DAl distributions. The distribution of Fe in seawater is controlled by rate of Fe stabilisation (formation of organic binding ligands) and Fe removal processes (Hopwood *et al.,* 2020; Shaked *et al.,* 2020). Marine organism (*i.e.*

phytoplankton) will produce organic ligands to enable DFe uptake for their biological processes (Shaked *et al.,* 2020; Ventura *et al.,* 2021). Thus, DFe concentration in coastal water and open ocean was strongly influenced by existence of natural iron-complexing organic ligands as > 99% of chemical speciation of dissolved Fe dominated by organic complexes (Shaked *et al.,* 2020; Mohamed *et al.,* 2011). Table 3 highlights the Fe speciation data obtained from 3 stations

Table 3. Fe speciation data for selected stations at three different areas

Station	Area	Depth (m)	[DFe] (µM)	[L_T] (µM)	[L'] (µM)	Log K (mol[1])	[LT]/[DFe]	FeL (%)
48	I	3	1.08	1.05	0.00	22.43	0.97	97.1
108	II	3	1.07	1.13	0.06	23.32	1.05	105.0
130	III	3	3.21	3.20	0.00	24.33	1.00	99.8

[DFe] dissolved Fe concentration; [L_T] total binding ligand; [L'] free iron binding-ligand; Log K stability constant of Fe ligand

representing each study area. Ligand concentration and ligand stability constant pattern are: Area III > Area II > Area I. Several studies on iron(III) speciation reported that generally [L_T] was higher than DFe in surface water either in coastal water or open ocean (Cullen *et al.,* 2006; Boye *et al.,* 2001, 2005; Croot *et al.,* 2004), as observed at Area II. Excess organic ligands were produced by phytoplankton in ways to prevent precipitation of DFe enabling it to be biologically uptake. However, excess concentration of DFe was observed at Areas I and III resembled result of study conducted at Sulu Sea and Celebes Sea by Kondo *et al.* (2007). DFe concentration at Sulu and Celebes Seas were from 0.46 nM to 1.48 nM while L_T range obtained was from 0.43 to 1.09 nM, respectively. Excess DFe in surface water (DFe > L_T) occurred due to several possibilities including the present of DFe iron hydroxide particles (colloidal Fe). Besides that, organic or/and inorganic complexes of DFe with weak ligands might not be detected during the analysis due to limitation of our method (Mohamed *et al.,* 2011). For Area I, DFe may also supplied from coastal via riverine input supported by reported value of salinity at Station 48 which was lower compared to Stations 108 and 130 (Idrus *et al.,* 2016). Area III was analysed with highest ligand concentration (L_T: 3.20 µM) and strongest organic ligands (log k: 24.33). Production process of organic ligands could differ between different areas controlled by sources of organic ligands itself. Possible sources of organic ligands in Area III were from atmospheric dust input either via dry or wet deposition. In addition, highest average concentration of Chl-*a* was analysed at Area III. Organic ligand was also biologically produced by bacteria and phytoplankton (Blanco-Ameijeiras *et al.,* 2020; Ardiningsih *et al.,* 2021) as well as during the plankton cell lysis (Poorvin *et al.,* 2004). Thus, high abundance of Chl-*a* at Area III could support the production of more strong class of

organic ligands than Area I and Area II. The stability constant range measured in our study (log K; 22.43-24.33) was higher than Kondo *et al.* (2007), at different area with the same application method. They reported that log K obtained at Sulu Sea, Philippine Sea, Celebes Sea, and South China Sea was 22.4 to 22.6, 23.3, 23.0 and 22.0, respectively. Log K obtained at coastal and open ocean water was 19.5-22.3 and 21.4-22.7, respectively (Mohamed *et al.,* 2011; Cullen *et al.,* 2006; Croot *et al.,* 2004; Croot & Johansson, 2000). However, there were some locations that analysed with strong Fe organic ligands such as Sulu and adjacent Seas (log K 22.0-24.0), Southern Ocean and equatorial Pacific Ocean (log K > 23) (Kondo *et al.*, 2007; Nolting *et al.*, 1998; Rue & Bruland, 1995), due to the intensidied light exposure and located at the shelf (Ardiningsih *et al.,* 2021).

CONCLUSION

This study has provided information on the dust deposition, atmospheric of total and dissolved Fe and Al fluxes, dissolved Fe and Al fluxes in the surface waters, and Fe ligands in the southern part of SCS (Sarawak waters, Malaysia Borneo) for the first time after ~ 20 years. Higher atmospheric of total Fe and Al fluxes compared to dissolved Fe and Al fluxes indicated that dust particles from the haze were the main sources of those trace metals at the study areas. Moreover, the dissolved Fe and Al fluxes on the surface waters from the nearby lands were considerably low. Al distribution data suggested that atmospheric dust deposition significantly enhance propagation of phytoplankton biomass along with Fe uptake. The uptake of Fe by phytoplankton possibly via production of natural organic Fe(III) binding ligands. Future studies should focus on understanding the role of natural organic Fe(III) binding ligands in this area along with phytoplankton analysis.

ACKNOWLEDGEMENTS

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