More surprises in the global greenhouse: Human health impacts from recent toxic marine aerosol formations, due to centennial alterations of world-wide coastal food webs.

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ABSTRACT

Reductions of zooplankton biomasses and grazing pressures were observed during overfishing-induced trophic cascades and concurrent oil spills at global scales. Recent phytoplankton increments followed, once Fe-, P-, and N-nutrient limitations of commensal diazotrophs and dinoflagellates were also eliminated by respective human desertification, deforestation, and eutrophication during climate changes. Si-limitation of diatoms instead ensued during these last anthropogenic perturbations of agricultural effluents and sewage loadings. Consequently, ~15% of total world-wide annual asthma trigger responses, i.e. amounting to ~45 million adjacent humans during 2004, resulted from brevetoxin and palytoxin poisons in aerosol forms of western boundary current origins. They were denoted by greater global harmful algal bloom [HAB] abundances and breathing attacks among sea-side children during prior decadal surveys of asthma prevalence, compiled here in ten paired shelf ecosystems of western and eutrophied boundary currents. Since 1965, such inferred onshore fluxes of aerosolized DOC poisons of HABs may have served as additional wind-borne organic carriers of toxic marine MeHg, phthalate, and DDT/DDE vectors, traced by radio-iodine isotopes to potentially elicit carcinomas. During these exchanges, as much as 40% of mercury poisonings may instead have been effected by inhalation of collateral HAB-carried marine neurotoxic aerosols of MeHg, not just from eating marine fish. Health impacts in some areas were additional asthma and pneumonia episodes, as well as endocrine disruptions among the same adjacent humans, with known large local rates of thyroid cancers, physician-diagnosed pulmonary problems, and ubiquitous high indices of mercury in hair, pesticides in breast milk, and phthalates in urine.

KEY WORDS

aerosols, biogeochemical cycles, marine systems, health impacts

HIGHLIGHTS

Oil spills, heavy metals, and overfishing decimated zooplankton grazers

Desert expansions and eutrophication concurrently fueled diazotrophs, not diatoms

Nitrogen-fixers set free commensal ungrazed dinoflagellate HABs

HAB increments caused ~45 million global asthma attacks during 2004

HAB aerosols served as surrogates of inhaled MeHg, phthalate, and DDT neurotoxins

Downwind of nuclear facilities radio-iodine carried by HABs also caused cancers

1. INTRODUCTION

Marine aerosols from harmful algal blooms [HABs] of *Karenia* and *Ostreopsis* spp. have been a persistent human health issue in coastal communities of the Gulf of Mexico [GOM]. Breathing difficulties during coincident fish kills had been first described off Vera Cruz, Mexico in October 1876 (Nunez Ortega, 1879; Magana et al., 2003). On the WFS [West Florida shelf], results from a complex zooplankton model, ZOOSIM [zooplankton simulations] on the top-down impacts of herbivore reductions during the last half-century demonstrated that wind-borne food web exports of aerosolized HAB asthma triggers still caused pulmonary distresses 140 years later. Yet, we had hypothesized that they were now co-transported with both other marine mercury (Walsh et al., 2015, 2016) and legacy pesticide (Stemmler and Lemel, 2009) neurotoxins, entained within sea salts and recorded as pulmonary illnesses among adjacent humans (Figs. 1-2).

Sea spray, as chlorine measured in rain water, had extended ~1200 km inland from the GOM to Indiana in July-September 1955 (Junge and Gustafson, 1957), during 13 tropical cyclones that year. But, small HABs prevailed on the WFS during this decade of 1955-1965, before onset (Fig. 3) of a trophic cascade on both sides of the GOM (Walsh and Steidinger, 2001; Walsh et al., 2011). Subsequently, between 1967 and 2011, increased HAB onshore wind-borne aerosol fluxes of halides and DOC-transported organic neurotoxins amounted to ~48% of annual carbon sequestration by ungrazed WFS HABs (Walsh et al., 2016). Incremental prevalences of asthma also occurred within downstream Indiana, from 4.9% in 1966 to 11.0% in 2003 (Arebiter, 1967; Akinbami et al., 2009).

During 2003, the GOM may still have remained a source of legacy DDE pesticide metabolites for dispersal to Indiana (Ho and Hites 2004), but via landfalls abetted by the

dinoflagellate-dominated HAB carriers along the Florida Pandhandle, rather than past the diatom-rich Mississippi River Delta (Fig. 2). Earlier DDE maxima in Alabama air during June and September 1996 (Jantunen et al., 2000) may have also reflected seasonal onshore fluxes of HAB-modulated neurotoxins of: **1**) other phthalate endocrine disruptors; **2**), organic mercury poisons; and **3**) asthma triggers during their usual peak summer-fall accumulations of dinoflagellate biomasses and subsequent lysed DOC carriers of lipophillic toxins.

During that two-year event, which began in September 1994 and ended by May 1996, until another HAB started in September 1997, a total of 238 manatees, ~10% of the Florida population, died from inhaling HABs. Such continued aerial imports of marine poisons were reflected in 2007 as part of recorded human asthma hospitalization rates over the SE United States, with corollary deposition of co-travelling total mercury aerosols in surface soils (Fig. 2) since 1965 (Walsh et al., 2015).

Over the past 50 years, global decimations of marine zooplankton herbivore populations (Table 1) were partly due to poisonings by petroleum (Almeda et al., 2013), DDT/DDE (Kannan and Sen Gupta, 1987), mercury (Hook and Fisher, 2001), phthalate ester (Price et al., 1986), and radionuclide (Jeffree et al., 1997) toxins. These population decrements of copepod herbivores were also the result of world-wide alterations of top-down controls via trophic cascades, caused by overfishing (Pauly et al., 1998; Walsh et al., 2011; 2015; 2016). Seafood consumption was a major past source of methyl mercury [MeHg] for humans (Sheehan et al., 2014). However, ubiquitous imprudent reductions of piscivore fish stocks may have shifted the importance of toxic marine Hg vectors from vertebrates to plankton, with present marine pollutant exposures to humans manifested as additional aerosol transports of organic and heavy metal poisons from coastal seas, traced by total halides and some of their radionuclides.

1.1 Top-down and bottom-up controls

Consequently, ~15% of total world-wide annual asthma trigger responses (Table 2), i.e. amounting to ~45 million adjacent humans during 2004, were due to marine brevetoxin and palytoxin organic poisons in aerosol forms of initial coastal dinoflagellate origins. Phalates also impacted humans by both inhalation and ingestion. By contrast, after eating contaminated reef herbivores and carnivores, e.g. parrotfishes and barracudas, ciguatoxins of *Gambierdiscus* spp. origins now poison about 250,000 people each year (Quod and Turquet, 1996), with few reported fatalities. Earlier, during inferred warmer periods of ~1000-1450, larger temperatures and greater incidences of ciguatera may then have prompted Polynesian emigrations away from the Cook Islands (Rongo et al., 2009), where a Biritish sailor was poisoned in 1774.

The more recent aerial vectors of pelagic and tychopelagic HABs had been overlooked by most environmental scientists, physicians, and economists, including prior studies of earlier *surprises* in inorganic ocean chemistry (Broecker, 1987). These causal interactions were reflected in greater global (Fig. 4) HAB abundances and associated breathing attacks among sea-side children and adults during 258 multi-decadal surveys of asthma prevalence during mainly the International Study of Asthma and Allergies in Childhood [ISAAC] program. We compiled some of these results as a few examples of asthma prevalence among humans, living next to ten paired shelf ecosystems of oligotrophic western [WBC] and eutrophied [EBC] boundary currents of coastal seas (Table 2).

The former nutrient-poor systems, adjacent to some deserts Fig. 4), were dominated by commensal pairs of diazotrophs and dinoflagellate fish-killer HABs. While, the latter nutrient-rich systems were fueled by coastal upwelling and river runoff to instead mainly grow diatoms. They had supported, in turn, food chains of large fisheries as protein sources for developing and developed nations. The successful competition of diazotroph-fueled dinoflagellate HABs against

silicate-limited diatoms partly followed altered desertification, deforestation, and sewage loadinges of most nutrients to coastal seas, except for silicon. In combination with decimation of copepod herbivores there as a dominant feature of world-wide overfishing-induced trophic cascades during overlapping time periods (Tables 1-2), a cumulative malign public health surprise indeed emerged over the last century.

The success of these non-diatomaceous phytoplankton led to wind-borne, aerosol-forced asthma and pneumonia episodes (Fig. 1), organic mercury poisonings, DDT/DDE and phthalate endocrine disruptions among coastal human populations within >300 km inland from global sea shores (Fig. 4). Where dissolved marine radio-iodine triggers of nuclear power and waste recycling plants were present, downstream sea sprays and air-borne HAB carriers evidently led to thyroid carcinomas as well.

We began this review, with consideration of the evidence for arrivals of onshore marine aerosol sources of illnesses, and deaths in some cases, from the GOM and South Atlantic Bight [SAB] of North America over the last century. We then placed these data in a holistic global context of the same phenomena in other analogs of South American [Brazilian and Chilean Seas], European [North and Mediterranean Seas], Indian [West Indian shelf, Bay of Bengal], African [Mozambique and South Africa shelves], Asian [Sea of Japan, Bohai, Yellow, East and South China Seas], and Oceania [French Polynesian, Coral, and Tasman Seas] coastal waters (Table 2).

Such inferred onshore fluxes of aerosolized HAB dissolved organic carbon [DOC] poisons (Walsh et al., 2015; 2016) may have also served as additional wind-borne organic carriers (Fagnani et al., 2012) of more neurotoxic marine methylmercury [MeHg], dichlorodiphenyldichloroethane [DDT], dichlorodiphenyldichloroethylene [DDE], and phthalate vectors to potentially elicit both other mercury poisonings and endocrine disruptions among the

same adjacent human populations. They were concurrently related to *known knowns* (Rumsfeld, 2011) of elevated seashore: **1**) asthma prevalences; **2**) DDT/DDE concentrations in human breast milk; **3**) MeHg hair indices of heavy metal poisons (Table 2); **4**) phthalate metabolites in urine; and **5**) thyroid carcinomas, where sea sprays and air-borne HAB poison carriers near coastal nuclear facilities moved downwind for radio-iodine mutagens to be inhaled by humans.

Once bottom-up Fe-, N- and P-limitations of commensal diazotrophs and dinoflagellates, as well as prior ten-fold greater top-down grazing demands (Walsh, 1976; Batchelder et al., 2012) of marine copepod herbivores (Table 1), were alleviated, neurotoxic HABs blossomed. These food chain intermediaries and their diatom competitors had previously passed bioaccumulated MeHg up marine food webs (Table 2) as just one set of ingested toxin pathways for trophic transfers to adjacent humans. Another set, carried within HAB DOC, was aerial dispersions (Walsh et al., 2015; 2016) of multiple organic poisons for direct inhalation by more sea-side victims.

1.2 Plankton dichotomies

A decline of copepod herbivores (Table 1) resulted in loss of their fecal pellets as vectors of carbon transport to the sea bottom. On the WFS between 2001 and 2010, this process coincided with 3-5 fold increases of lytic algicidal bacterioplankton activity against HABs (Lenes et al., 2013; Walsh et al., 2016). Replacement of herbivore zooplankton grazing of phytoplankton biomass by lytic bacterioplankton was reflected in decadal increases of WFS summer microbial-recycled stocks of ammonium throughout the water column (Walsh et al., 2016). Concomitantly, a shift in loci of HAB lytic losses and nutrient recycling by WFS microbial heterotrophs occurred, moving from not just the hypoxic sea floors, but also to near surface waters. Alteration of these heterotrophic processses, from copepods to microbes,

facilitated mobilization of organic toxins from the WFS, entrained as lysed dissolved organic carbon [DOC] within surface sea spray aerosols, ejected by winds and shoaling waves.

During photorespiration to cope with excess light, diatoms released small sugars like glycolic acid (Fogg, 1983), but few marine bacteria have evolved to harvest this carbon source (Lau and Armbrust, 2006). Whereas in contrast, shade-adapted, pelagic migratory *Karenia mikimotoi* and *K. brevis* did not excrete this acid, using it instead as a building block during formations of their respective neurotoxins, i.e. various gymnocin and brevetoxin poisons (Satake et al., 2002; 2005; Chattopadhyay et al., 2007; Van Dolah et al., 2009). The same polyketide synthesis pathway was also evidently employed by the benthic, very toxic dinoflagellates *Gambierdiscus* spp. (Pawlowiez et al., 2014) and *Ostreopsis* spp. (Ramos and Vasconcelos, 2010), implicated in ciguatera poisoning events, to produce their respective ciguatoxins, maitotoxins, and palytoxins. But, we suggest here that the latter tychopelagic dinoflagellate is an unlikely ciguatoxin vector to humans, compared to that of former benthic HAB via demersal fish herbivores.

The malign impacts of these neurotoxins varied greatly, at least with respect to laboratory mice. Like the emergence of prior cryptic HABs of *Ostreopsis lenticularis* off Puerto Rico by 1985 (Ballantine et al., 1988), HABs of *Ostreopsis heptagona* appeared in the Florida Keys during the same year (Norris et al., 1985) and off Veracruz by 2005 (Okolodkov et al., 2007). In terms of poison potency, both *Ostreopsis ovata* and *Gambierdiscus toxicus* were ~4 million-fold more poisonous to small mammals than *K. brevis*. The lethal dosage at which half the mouse assay population died [LD_{50}] from palytoxins of *O. ovata* was 0.045 µg kg^{-1} (Ramos and Vasconcelos, 2010) and 0.050 µg kg^{-1} for maitotoxins of *G. toxicus* (Murata et al. 1993),

compared to a much larger LD_{50} of 180 mg kg^{-1} for brevetoxins of *K. brevis* (Lin et al., 1980; Shimizu et al., 1986).

To be successful, each phytoplankton species must either occupy a prior open niche (Hutchinson, 1965), or compete positively against the present occupant. As pelagic predators, *Karenia* spp. kill fish to harvest supplemental nutrient supplies from positively buoyant decaying fish within the euphotic zone, in addition to utilization of ambient nutrients from the water column and those released by vertically co-migrating *Trichodemium* colonies (Mulholland et al., 2006; Walsh et al., 2006; 2009) – all fueling the nitrogen demands of concurrent photosyntheis.

Whereas, *Ostreosis* spp. are tychopelagic, spending time both in the water and as an epiphyte on hard coral sea bottoms and macrophytes, evidently at times possibly consuming sea urchins under a muscilaginous blanket (Shears and Ross, 2009; Graneli et al., 2011; Amzil et al., 2012; Pezzolesi et al., 2014). However, the nutritional value to *Ostreospis* spp. from poisoned sea urchin juveniles (Privitera et al., 2012) is presently unknown, so it may simply be a HAB defense posture against those toxic, in turn, echinoderm hebivores. None of the HAB playtoxins have been found within parrotfish herbivores (Suzuki et al., 2013), so presumably these dinoflagelellate mixotrophs also do not harvest demersal fishes.

In contrast, *Gambierdiscus* spp. usually represent a benthic shunt of element cycling, since thay are rarely found in the overlying water column, unless the sediments are physically disturbed (Rougerie and Bagnis, 1992) by military constructions (Ruff, 1989), other practice bombings (Porter et al., 2011), and storms (Skinner et al., 2011). They produce mucous as well, but again their consequent toxicological and ecological impacts are unknown (Zingone, 2012). Presumably in and/or near the sediments, *Gambierdiscus* spp. were not subject to nutrient-limitations, because of usual large interstitial concentrations of nitrate and phosphate.

Using estimated half-saturation constants of 0.39 umol NO_3 l^{-1} and 0.03 umol PO_4 l^{-1} in a one-dimensional model of Hawaian ciguatera agents, the simulation results replicated observations of both *Gambierdiscus* spp. and near-bottom nutrients of the shallow water column (Parsons et al., 2010). Within deeper water columns, commensal *K. brevis*, as benificiaries of nutrients released from *Trichodesmium* spp. instead had simulated half-satuation constants of 0.50 umol NO_3 l^{-1} and 0.20 umol PO_4 l^{-1} (Walsh et al., 2001), to cope in models and "the real world" with the oligotrophic habitat of those shelves, where they live adjacent to western boundary currents. Similarly, these latter kinetic parameters of nutrient uptake of dissolved nitrogen and phosphorus were the same in a recent model of the production of toxins by *O. ovata* (Pinna et al., 2015).

Nevertheless, the respective maximal realized specific growth rates of field and laboratory populations of *K. brevis* (Walsh et al., 2001), *O. ovata* (Vidyarathna and Graneli, 2012; Scalco et al., 2012) and *Gambierdiscus* spp. (Kibler et al., 2012) were different values of 0.8 day^{-1}, 0.3 day^{-1}, and 0.2 day^{-1} at 30^{o}C. They also have overlapping light-saturations of 65 (Walsh et al., 2001; Magana and Villareal, 2006), 50-100 (Scalco et al., 2012; Monti and Cecchin, 2012), and finally 35-110 (Kibler et al., 2012; Xu et al., 2016) µmol photons m^{-2} sec^{-1}, or = µE m^{-2} sec^{-1}. Thus, their HAB niche separations may be maintained more by physical habitat dislocations of water and sediments, with different fluxes of nutrients and light at the vertical boundaries, rather than by just these minimally varying HAB physiological properties, except for their divergent intrinsic growth rates.

In contrast, at 30^{o}C, the diatom *Skeletonema costatum* has a growth rate of 2.4 day^{-1} under a light saturation intensity of 190 µmol photons m^{-2} sec^{-1}, compared to 0.8 day^{-1} and 300 µmol photons m^{-2} sec^{-1} for *Trichodesmium* (Walsh et al., 2001), i.e. they are not occupants of the

"dark side" of a HAB world. Of course, organic nutrient sources of poisoned prey, and grazing losses to copepod, parrotfish, and mollusk herbivores (Table 1) – as discussed later – complicate simple model paradigms. Causal numerical models must now be revised to consider perturbed coastal systems of more than one resident HAB. Furthermore, given these insights, future requirements of cross-disciplinary eco-medical consiliences (Wilson, 1998; Pauly, 2001) of both environmental scientists and physicians are necessary for solutions to compounded, century-old problems of multiple imprudent human uses of natural resources, which have together allowed global HABs to expand.

Moreover, recycled DOC supplies from diatoms come mainly in the form of "sloppy" grazing by copepods depending upon the size of algal prey, not usually from their excretions, ranging from 20% to 62% of their inept consumption of phytoplankton fixed carbon (Moller and Nielsen, 2001; Saba et al., 2011). Thus, as a predation strategy among lower trophic levels of the GOM, and presumably elsewhere, dinoflagellates are more easily lysed there by viruses (Ortmann et al., 2011) and bacteria (Douchette et al. 1999), than are the diatoms. Most unfortunately, when each of these dinoflagellate HABs of *Karenia* and *Ostreopsis* spp. are in the water column, they form aerosols, as part of their lysed DOC within sea sprays along the shorelines. Whereas, at least *Gambierdiscus* spp. of the benthos apparently do not directly form organic aerosols, but their ciguatoxins may still be entrained within those of the other ambient HAB organic carriers, upon sediment disruptions.

Accordingly, a greenhouse surprise of more successful diazotrophs and commensal dinoflagellates, in global competition with diatoms after human desertification, deforestation, eutrophication, and overfishing in trophic cascades over the last 100 years, did not bode well for adjacent humans (Table 2). Dispersion of marine pelagic and epibenthic HAB poisons and their

co-travelling pools of sea salts and radio-iodine tracers, together with other MeHg, phthatlate, and DDT neurotoxins, had particularly spread among those humans, who lived inland within at least one thousand kilometers of western boundary currents [WBCs]. Ciguatoxin HAB carriers of *Gambierdiscus* spp., eaten mainly by reef fishes, instead remain in the vicinity of surviving coral platforms, depending upon local currents and sediment resuspensions for propagule exports.

Recall that the "winnowing oar" of Homer's *Odyssey* was when the ancient Greeks were supposed to walk far enough inland during ~800 BC, until an oar was mistaken to be a threshing device. The downstream "winnowing marine HABs" of today would similarly serve as wind tell-tales, when and where they rained out to no longer plague adjacent humans in their inhalant forms, at sufficient distances away from world-wide sea shores.

Unfortunately, insecticide aerosols from subtropical and boreal seas have also spread (Semeena and Lammel, 2005; 2009), along with other past cesium and iodine radiotracers of the Chernobyl and Fukushima Dai-ichi accidents (Cristoudias et al., 2014), to polar regions (Cone, 2005). There no pesticides were applied, nor phthalates synthesized, locally. They instead moved north via both WBCs and in a faster atmospheric, temperature-dependent *grasshopper* distillation process (Wania and Mackay, 1993).

En route within underlying surficial hypoxic sediments, both DDT was metabolized to DDE (Wedemeyer, 1967) and sulfate-reducing bacteria effected methylation conversions of inorganic mercury to MeHg (King et al., 1999). Our hypothesized coastal biophysical pollutant distillation [CBPD] processes within WBCs, introduced here, would thus have also occurred repeatedly beneath aerial *grasshopper* transport events to concentrate marine toxins. For example, within the Loop Current on the Louisiana shelf, storm-induced sediment resuspensions of temporarily-stored marine neurotoxins, e.g. mercury at mid-shelf, were observed (Liu et al., 2009). Once air-borne (Wania and Mackay, 1993), onshore and poleward transports of renewed

marine organic pollutants and their radio-tracers would both be continued by wind-wafted (Walsh et al., 2015) HAB carriers (Fig. 2) and be consistent with predicted ~2-fold faster poleward shifts of marine dinoflagellates, than those of diatoms (Barton et al., 2016).

1.3 HAB carriers

The accompanying marine exports of these neurotoxins and their radioactive tracers were facilitated by multiple DOC carriers (Duce et al., 1963; Blanchard, 1975; Fagnani et al., 2012), including those of bacterial-lysed phytoplankton origins (Doucette et al, 1999; Roth et al., 2007, 2008a,b; Lenes et al., 2013). Early studies led to measurements of ~20-fold iodine enrichment within atmospheric sea-salt particles, compared to dissolved halides and their radionuclides left behind in seawater (Seto and Duce, 1972). Subsequent experiments found the same ~20-fold enrichment of brevetoxins of *K. brevis* = *Ptychodiscus brevis* within their overlying air aerosols, in contrast to those of surface sea water (Pierce et al., 1990). Later, MeHg compounds were observed to be more soluble in DOC carriers, as well (Fagnani et al., 2012).

2.0 Gulf of Mexico and downstream South Atlantic Bight pollutant perturbations

Despite prior warnings of potential recycled emissions of legacy organic mercury neurotoxins from temporary storage pools of coastal seas, amounting to ~89% of the total Hg effluxes across the sea-air interface (Strode et al., 2007; Amos et al., 2013), their public health impacts, compared to only eating fish (Sheehan et al., 2014), have been neglected. Similarly, the fates of known mercury aerosols, found in air above the Florida Panhandle and adjacent Mississippi (Engle et al., 2008; Rolison et al., 2013; Ren et al., 2014), compared to those accumulated in surface soils (Fig. 2), were also mainly ignored.

We color coded (Fig. 2) the Centers for Disease Control [C.D.C.] reports of deaths, due to pneumonia and influenza, within 1027 Counties of the SE U.S. during 1999-2011. Overlain on this map (Fig. 2) is our additional linear interpolation of both 250 United States Geological Suvey [U.S.G.S.] surface soil measurements of total mercury [THg], taken ~80 km apart during 1961-1970 (Shacklette et al., 1971), and 45 core samples obtained druing 1992 in the Florida Everglades (Rood et al. 1995), shown as the open black square symbols of Figure 2.

Note that the prior mean mercury soil content of the Florida Everglades was ~41 ng THg g^{-1} [ppb] in 1970, dated from these soil cores by ^{210}Pb and ^{137}Cs chronologies (Rood et al. 1995). This extrapolated down-core estimate is an intermediate value of the measured SE U.S. mercury contamination field during the 1960s (Fig. 2). Maximal human mortalities of >50 deaths per 100,000 residents (Fig. 2) from influenza and pneumonia reported by the Centers for Disease Control [CDC] within 1027 Counties of the SE U.S. during 1999-2011 were co-located spatially with large accumulations of >150 ppb of total mercury within surface soils (the red isopleth of Fig. 2), albeit in different years within the limits of available data sets.

When air-borne marine asthma triggers of sea spray-entrained HABs above the United States penetrated past Georgia (Fig. 2), as far inland as Indiana ((Junge and Gustafson, 1957), they carried along both MeHg and DDT neurotoxins, while serving as an example of similar onshore global phenomena. For example, residents of both coastal and inland states of the U.S., who did not eat fish, nevertheless had total mercury hair concentrations, ranging from 0.01 to 0.20 ug THg g^{-1} [ppm] during 1998-1999 (Knobeloch et al., 2005). Moreover, among Swedish inhabitants that also did not consume fish, a somewhat larger range of 0.04-0.32 ug THg g^{-1} was found in human hair (Lindberg et al., 2004). We shall find that in other regions, measured hair mercury contents of proclaimed vegetarians amounted to ~40% of those residents, who

consumed both plants and fishes, due to inhalation of Hg aerosols by the former, and both breathing and eating by the latter.

These greater values of some accumulated European mercury poisons in hair perhaps reflected both biotic transformations of mercuric slimicides, used in Scandinavian pulp factories, and shorter downstream distances of HAB DOC carriers of multiple organic neurotoxins (Fagnani et al., 2012), from marine boundaries of the Baltic Sea. In Norwegian coastal forests, impacts of winter sea sprays were found at ~40 km inland distances from near shore marine waters (Aamlind and Horntvedt, 2002). Furthermore, independent budgets of Hg loadings to residents of Senegal in West Africa (Niane et al., 2015) and of Cambodia in East Asia (Agusa et al., 2005) concluded that other mercury sources, besides those of fishes, must not be ignored there, as well.

The black contour line of Figure 2 designated a soil concentration of only 15 ppb THg, compared to the red contour of 150 ppb. Thus, maximal human mortalities of >30 deaths per 100,000 residents (Fig. 2) from influenza and pneumonia reported by the CDC during 1999-2011 were co-located with large accumulations of total mercury within surface soils, albeit in different years. At times, pre-existing asthma conditions had also exacerbated pneumonia infections (Palmer et al., 2003, 2009; Klemets et al., 2010; Toren et al., 2011), such that we hypothesized the spatial pattern of influenza and pneumonia deaths (Fig. 2) was also an index of morbidities, derived from air-borne, onshore transports (Walsh et al., 2015; 2016) of marine HAB asthma triggers.

Similar to 19th century colonial episodes (Nunez Ortega, 1879) of human pulmonary morbidities and mortalities, lung maladies recently became more frequent phenomena along both Texas and Florida (Fig. 1) coastlines (Cheng et al., 2005a,b) of the GOM (Fig. 2). Reports of

inhalation of HAB aerosols increased along the Florida west coast, after the 1940s (Woodcock, 1948; Kirkpatrick et al., 2006, 2010, 2011). During 1978-1983, human respiratory deaths in Florida (Fig. 1) and pelagic HAB abundances along both the WFS and Campeche Bank of Mexico (Figs. 2a, b) had all increased ~3-fold, i.e. those near-surface phytoplankton seen by satellites.

2.1 GOM radiotracer of thyroid cacinomas and pesticide endocrine disruptors

A corollary of this hypothesis was that due to both co-travelling, wind-borne methyl mercury [MeHg] and DDT/DDE aerosols, the repeated spatial pattern of THg remaining in soils (Fig. 2) presumably reflected additional onshore transports of sea spray and other organic neurotoxins, in addition to those of the HAB carriers. The heavy metal would have been methylated in nearshore GOM sediments (King et al., 1999), while DDT would no longer just be stored in shelf depocenters, with inland transfers of both neurotoxins traced by radio-iodine isotopes. Indeed, the annual rate of increase of thyroid cancer tracers during the 1980s in Florida (Mullu and Margo, 2000) was also the same as that of the human asthma (Fig. 1) and HAB increments (Fig. 3). But, the source of this radionuclide in the GOM was mainly radio-iodine fallout from U.S. bomb tests, not from point sources of local nuclear power and waste recycling plants.

Exceptions were two power plants: the boiling water reactors at *Laguna Verde* opened during 1990 in Veracruz, Mexico; and the recently closed *Crystal River*, next to the WFS. Instead, the *Turkey Point* nuclear reactors east of Homestead, Florida drained into the SAB. Among the 32 States of Mexico, Veracruz had the third highest rate of child-hood cancers of <15 years age during 1980-1995, including those of acute lymphoblastic leukemia [ALL]. It increased 3-fold between 1982 and 1991 (Abdullaev et al., 2000). Greater mortalities due to ALL

bone cancers are compounded by pneumonia infections, which may have followed arrival of wind-borne asthma triggers from the GOM to young residents of Veracruz.

In terms of far-field radio-iodine tracers, the ~100 atmospheric bomb tests above the Nevada Proving Grounds, during the 1950s at 105 km NW of Las Vegas, still left behind wind-borne ^{129}I signals in the Mississippi Delta sediments during this time period, after core collections made in 1993 (Oktay et al., 2000). These air-borne influxes of ^{129}I were then also traced farther downstream in the form of radioactive thyroids of free-ranging cattle in Florida (Van Middlesworth, 1954; Marston, 1958). Finally, since DDT aerosols were also the same size (Giles et al., 1995), these metabolized prior pesticides, previously used in crop management, public health applications, and now stored in marine sediments, should have moved inland, as well.

The GOM sediment legacy concentrations of this pesticide remained high in most coastal depocenters. Indeed, the DDT stocks of *Crassostrea virginica* increased by 1992 within mollusk indices of surface sediments at the 60-m isobath on the mid-shelf of the Mississipi River Delta (Santschi et al., 2001). Yet, the DDT concentrations of these oysters were an order of magnitude greater in Tampa Bay of the WFS (Santschi et al., 2001), although ten-fold smaller supplies of freshwater loadings occurred there (Gilbes et al., 1996). Nevertheless, HAB carriers of organic nuerotoxins were also very abundant on the WFS, compared to the diatom habitat of the eutrophic Louisiana shelf, on the western side of that phytoplankton ecotone, along the cross-shelf DeSoto Canyon (Walsh et al. 2006; 2015).

2.2 Sea spray aerosol emissions

The black contour line of Figure 2 designates a soil concentration of 15 ppb THg [15 ng THg g^{-1}], compared to the red contour level of 150 ppb. Together, these composite data

(Shacklette et al., 1971; Rood et al. 1995) depicted little inland soil contamination of <15 ng THg g^{-1} dw, past Tampa Bay to the northeast. By contrast, ten-fold greater Hg-contaminated, surface soils of >150 ng Hg g^{-1} dw occurred along the Florida Panhandle, where wave heights during 1970 were larger than those off central West Florida.

Within the Big Bend region (Fig. 2) of the WFS, 130 km south of Apalachicola at NOAA buoy NDBC 42036, the observed wave heights were ~3 fold greater during October 2007, than those measured at our moorings above the 10-m and 20-m isobaths off Sarasota, Florida. Since waves break within shallow waters, when wave heights are ~80% of the water depth, expulsions of malign aerosols in sea sprays were more likely to have occurred over a larger export region off the Florida Panhandle, where high mercury of both soil (Fig. 2) and human hair were noted.

2.3 Confirmatory hair mercury distributions

Mercury contents of human hair reflect both marine and terrestrial sources of organic mercury within their diet, as well as any inhalation of Hg aerosols. The WHO limit for ***acceptable*** total mercury concentration is 2.5 ug THg g^{-1} [ppm] for hair samples (Bellanger et al., 2013). Yet, among residents of Alabama, their hair mercury content was 2.8 ug THg g^{-1} in Alabama (Warner, 2007), near Mobile Bay and the Florida Panhandle, during 2007 at varying downstream distances from coastal chlor-alkali plants in Texas, Louisiana, and Alabama.

By contrast, the arithmetic mean of human hair mercury near Port St. Lucie in Martin County, ~1325 km downstream from Mobile via Loop Current and Gulf Stream WBCs along the west and east coasts of Florida, was a smaller amount of only 0.7 ug THg g^{-1} (Nair et al., 2014). It reflected less remaining soil mercury stocks of the initial onshore marine aerosol dispersals (Fig. 2), where HAB carriers were also usually rare (Walsh et al., 2009). Farther north within

Florida's Duval County in 2006, hair mercury declined again to 0.3 ug THg g^{-1} (Traynor et al., 2013).

However, additional industrial supplies of mercury contaminants were added to the SAB by Georgia commercial firms, with ~10-fold larger hair contents found among downstream human residents of South Carolina. Consquently, along the SE U.S. seaboard during 1989-2001, human hair contained a mean of 1.1 g THg g^{-1} (Table 2). In comparison, only 0.4 ug THg g^{-1} (Knobeloch et al., 2005) was found in the hair of California residents, living adjacent to the diatom-dominated eutrophied boundary currents [EBC] of the California Current system (Table 2). Furthermore, during 1999-2000, the arithmetic mean hair mercury content among all U.S. females was only 0.5 ug THg g^{-1} (McDowell et al., 2004). Here, EBCs are defined as nutrient-rich coastal habitats, due to either upwelling within eastern boundary currents, or riverine nutrient supplies, i.e. in contrast to oligotrophic WBCs.

Upon entrainment within expelled sea spray aerosols, dissolved brevetoxins previously lysed by bacteria at HAB demises, in addition to a DOC component from "sloppy feeding" by copepods, would exit coastal seas, carrying multiple other neurotoxins. Earlier inorganic Hg stocks would have been methylated to more poisonous organic forms, while DDT stocks were partly converted to other long-lived DDE metabolites. But, they would leave the sea surface as aerosols, after *in situ* biotic modifications, at very different time scales.

Sulfate-reducing bacteria quickly methylate inorganic mercury to MeHg of surficial sediments within 12 hours, such that these pore waters contain as much as 80% organic mercury, from Texas (Gill et al., 1999; Bloom et al., 1999) to Florida and Georgia (King et al., 1999, 2000). Whereas, dechlorinating bacterial conversions of predominate para isomers of DDT to the

DDE metabolites (Albone et al., 1972; Lal and Saxena, 1982) are much longer, with a mean DDT residence time of 11 years within sediments of Monterey Bay (Phillips et al., 1975).

2.4 Chronologies of pesticide metabolism on both sides of the GOM

Consequently, DDE/DDT ratios of marine biota provide a rough chronology of pesticide loadings and bacterial activities, with 10.4% of initial dech lorination effected by anaerobic bacteria, and just 1.6% by aerobic micro-organisms (Wedemeyer, 1967). In the Baltic Sea environs, where pesticides had been prohibited by ~1970, DDE/DDT ratios of human breast milk among residents of Finland and Denmark were 21.1 during 1997-2001 (Damgaard et al., 2006). Similarly, recall that, following publication of *Silent Spring* (Carson, 1962), DDT was also banned in the United States by 1973.

Yet, subsequent blood samples collected in 2003-2004 from 12-year old children in the United States, i.e. those born in 1991-1992 after a 19-20 year cessation of U.S. pesticide applications, nevertheless still contained a mean DDE content of ~105 ng g^{-1} lipid. These additional data possibly suggested consumption of imports of external contaminated foods from other countries, continuing to use pesticides on their agricultural crops (Patterson et al., 2009).

However, inhalation of prior terrestrial pesticide usage in the form of dust and other aerosols was a major DDT contamination route for humans (Ritter et al., 2011). Wind-borne onshore transports of legacy marine stores of pesticides in the GOM also thus can no longer be ignored, since recent aerial returns to humans of these co-travelling marine neurotoxic DDT poisons and their metabolites have become a global phenomenon (Stemmler and Lammel, 2009), like that of MeHg (Amos et al., 2013). Onshore fluxes were aided by both warmer surface temperatures for gas volatilization (Samuel and Pillai, 1989; Noyes et al., 2009) and growth of diazotroph-fed HAB carriers of organic toxins within oligotrophic WBCs.

We shall thus find here that world-wide health indices of recycled marine DDT in human breast milk, like those of hair mercury, urine endocrine disruptors, and inhaled asthma triggers (Table 2), had become joint wind-borne aerosol contaminations, teleconnected (Namaias, 1972) by atmospheric circulation fields and following, in turn, sea spray evasions from coastal shelves. These processes had one factor in common – expansion of African and Asian deserts, with downstream transports (Fig. 4) of incremental dust supplies of nutrients for *Trichodesmium* and other diazotrophs, which eventually led to HAB-modulated incremental outbreaks of asthma, hydragyria, endocrine disruption, and cancer.

Since both Saharan dust and *Trichodesmium* colonies were described off South America by Charles Darwin (1846), these are not new teleconnections. Instead in this review, we describe how in 20 coastal ecosystems (Table 2) physics and biology have conspired over the last century of anthropogenic pertubations to return eight sets of marine neurotoxins [brevetoxins, gymnocins, palytoxins, ciguatoxins, DDT, DDE, phthalates, and MeHg] from coastal seas to adjacent humans, with concurrent transport inferences drawn from dispersions of radio-chemical $^{129,\ 131\text{-}135}$I signals of half-lives, ranging from 15.7 million years to <60 days.

Given the pending public health, sociological, and economic consequences of such wind-borne poisons from coastal seas, enormous efforts have been devoted to collections of environmental and medical data. For example, along the northern GOM during 1986-1987, the mean DDE/DDT ratio of 301 surficial oyster samples was only 0.7 (Sericano et al., 1990), reflecting a recent pesticide source, since production of the DDE metabolite had been minimal in those few years.

When present, these same regions also had a mean MeHg/THg ratio of 15% in another 19 surface samples during 2006-2007 (Apeti et al., 2012), reflecting more active metabolic

processes of another group of anaerobic bacteria. During the intervening 1990s, 81% of the 37 northern GOM estuaries exhibited summer anoxia (Bricker et al., 1997), thus facilitating methylation activities of sulfate-reducing bacteria, as well as those of dechlorinating anaerobic bacteria (Wedemeyer, 1967).

This initial NOAA data set (Sericano et al., 1990) was collected after the first occurrence of a mean HAB of >100 ug chl l^{-1} in 1985 along the 5-m isobath off western Florida. It was ten-fold larger than the prior seasonal maxima during a continuous time series of weekly mean HAB stocks over ~50 years, compiled by us from 1965 to 2016 (Walsh et al., 2016). Under southerly winds of computed back trajectories and observed low radon contents, marine aerosols of sea salts, Saharan dusts, and organic particles of 1-10 microns sizes were also carried onshore near Houston, Texas in August-September 2006 (Bates et al., 2008), as part of the Texas onshore tongue of organic toxins from the GOM (Fig. 2).

Previously, within other near shore air masses of more southerly winds above the GOM in March-April 1977 between Louisiana and Texas, the DDE/DDT ratio of aerosols and gases was still 2.6 (Giam et al., 1980), suggesting then a relatively recent pesticide source in marine sediments, despite a 5-year ban on their usage, begun in January 1973. By contrast, in the air above agricultural cotton fields of Stoneville, Mississippi at ~415 km inland from the GOM during 1973, the DDE/DDT ratio had been ~0.1 (Arthur et al., 1976).

In October 2000 off Corpus Christi, Texas, brevetoxin aerosols of 7-9 microns diameters were also found in air (Cheng et al., 2005a). Thus, subsequent WFS HABs of very large biomass accumulations of >100 ug chl l^{-1} continued during 2001, 2005, and 2006, as a very large vector of onshore neurotoxic transfers from the northern GOM. They indicated that the transitions in the trophodynamics of this shelf ecosystem had previously occurred, before the *Deepwater Horizon*

oil spills exacerbated aerosol exports of organic neurotoxins, from an already unbalanced food web there and in the southern GOM (Walsh et al., 2015).

2.5 Southern GOM analog

Following the original description of breathing difficulties in Vera Cruz, Mexico during 1876 (Nunez Ortega, 1879), ecological and public health problems escalated over the next century, with the blow out of the *IXTOC* drilling platform on the Campeche shelf at ~19°24'N, 92°18'W (Walsh et al., 2015). This spill site was located southwest of the upstream rich fishing gounds, due to upwelling (Zavala-Hildago et al., 2003) on the Campeche Bank [22°18'N, 89°00'W]. The IXTOC spills released ~5.6 x 10^5 m^3 of crude oil there during June 1979 – March 1980 of another trophic cascade (Fig. 3c), similar to those petrochemical poison loadings from the *Deepwater Horizon* blow out, with subsequent effluxes of ~7.8 x 10^5 m^3 of crude oil during April-July 2010.

Consequently, local Bay of Campeche [BOC] copepods quickly died around the Mexican *IXTOC* well head, with 50% less zooplankton found on the Campeche shelf in June 1979, than previously in August 1978 (Guzman del Proo, 1986). By November 1979, zooplankton biomass at the *IXTOC* spill site had further declined to 13% of the stocks in August 1978 (Guzman del Proo, 1986). Subsequently, the ungrazed phytoplankton there during October-November 1980 (Fig. 3b) drifted downstream to Veracruz (Jernelov and Linden, 1981).

Set free from the grazing pressures of decimated zooplankton herbivores and entrained onshore within continued additional "sea sprays" (Sierra-Beltrain et al., 1998), post-IXTOC HABs moved towards the Mexican coast. More than 225 acute cases of childhood asthma episodes, presumably due to HAB air-borne triggers of *K. brevis,* were reported to the emergency

room of a Merida, Yucatan hospital at ~20°58'N, 89°37'W during September-October 2005 (Aguilar et al., 2009).

They were evidently the only aerosolized HABs present in the southeastern GOM, away from the eutrophic diatom-rich (Soto et al., 2014) upwelled waters (Zavala-Hildago et al., 2003) on Campeche Bank, that would have benefitted from oligotrophic diazotroph sources of nutrients, since no *Ostreopsis* spp. were observed there. In contrast, these more potent tychopelagic HABs persisted in Veracruz during 2008 (Okodlokov et al,, 2007; 2009). Previously, during 2002-2003 ~two-fold more asthma prevalences had also occurred among school children within downstream Tamaulipas State, than in upstream Yucatan State of Mexico (Lai et al., 2009).

During fall seasons in the SE GOM, the usual northerly onshore winds and high sea levels, i.e. piled up water at the coast, prevailed during the wet season (Fuentes-Yaco et al. 2001). Such a seasonal downwelling circulation pattern off Campeche would have effected nutrient-poor surface waters, favoring the commensal set of precursor diazotroph nutrient donors and ungrazed HAB beneficiaries. Earlier fall HABs of *K. brevis* along this GOM coast of Mexico had co-dominated the October phytoplankton community of the seasonal oligotrophic habitat with the nitrogen-fixer commensal nutrient donor *Trichodemium erythraeum*, over the prior half-century (Khromov, 1969; Sierra-Beltrain et al., 1998; Licea et al. 2004; Ake-Castillo, 2011).

As a result of their faster growth rates, local *K. brevis* HABs in Yucatan coastal waters were able to compete effectively against populations of *O. ovata*, despite reduction of grazing demands on both HABs. After onset in 1978 (Fig. 3c) of a parallel Mexican trophic casade (Walsh et al., 2011), the former pelagic *K. brevis* populations would no longer have been subject to larger copepod grazing stresses, once piscivore-freed increments of clupeoid zooplanktivore

sardines *Sardinella aurita* and herrings *Opisthonema oglinum* began to eat these crustacean herbivores. Subsequent fugitive IXTOC petrochemicals would have poisoned copepods as well.

By contrast, the grazing losses of the latter *Ostreopsis* spp. HABs must have also declined to smaller predatory demands of epibenthic herbivores along the Yucatan coast, once diseased sea urchin herbivores *Diadema antillarum* of upstream Panamanian origins had arrived off eastern Mexico by July 1983 (Lessios et al., 1984). These echinoderm hebivores only recovered to 10% of their prior stocks throughout the Caribbean Sea, as monitored at St. John, U.S. Virgin Islands by 2011 (Levitan et al., 2014).

Although the benthic HAB *Gambierdiscus toxicus* was found year round during 2008 at nearby seaside Chelem, Yucatan (Okodlokov et al,, 2007; 2009), nevertheless a 50% reduction of coral reef cover was observed between 1985 and 2005 among hard bottom habitats in this southeastern GOM region of the Mexican shelf (Harwell et al., 2007). A diminished spatial extent of their poison impacts was consistent with few episodes of ciguatera, noted there along the Caribbean Mexican coast during the 1980s and 1990s (Sierra-Beltrain et al., 1998). Moreover, the ecological and public health implications of these HAB separations in space along the southern GOM will soon be discussed here, in relation to similar separations in time. Off both Puerto Rico (Ballantine et al., 1988) and farther east within the British and U.S. Virgin Islands (McMillan et al., 1986; Tindall and Morton, 1998), dominance of first *Gambierdiscus* was followed by that of *Ostreopsis.*

In terms of more human health burdens from these GOM seas, a compounding onset of "fresh" pesticide loadings of possible transitory marine origins, due to other recent anthropogenic perturbations, can be inferred. These insecticides had also been recorded in sediments of the nearby sea-side Sabancuy Lagoon in Campeche, Mexico, after 2002. This study followed

extended Mexican usage of DDT to combat malaria and dengue fever until 1995, as well as removal of agricultural pests. Furthermore, that Mexican sediment DDE/DDT ratio of only 1.3 (Ramirez-Elias et al., 2016) was close to the value of 0.7, found in oysters of the northern GOM during 1985 (Sericano et al., 1990).

Thus, molluscan benthic herbivores do indeed serve as biotic sedimentary records of temporary pollutant storage pools in nearshore depocenters. Downstream within Laguna Mandinga off Veracruz in 2009, the same oyster pesticide DDE/DDT index of *C. virginica* again had a value of 1.0 (Longo-Reynoso et al., 2013). Such congruence suggested little difference between algal prey, consumed by these herbivores, and uneaten phytodetritus, left behind to accumulate within at least surface sediments.

Like in the northern GOM, this small Mexican DDE/DDT ratio was again presumably due to onshore imports of HAB carriers of brevetoxins and palytoxins, mainly unmetabolized DDT pesticides, and benthos-modified MeHg toxins. Once more, like on the WFS (Fig. 3a), satellite-estimated HAB biomass off Campeche in October 1980 was ten-fold greater than that seen by the same Coastal Zone Color Scanner [CZCS] there during October 1978 (Fig. 3b). Accordingly, more abundant HAB carriers were detected by the post-Ixtoc satellite imagery and observed by sickened residents to move downstream, from Campeche to Veracruz and Taumalipas (Jernelov and Linden, 1981), with later indications of hydragyria.

Finally, the mean hair mercury concentration of humans in upstream Columbia, living near an abandoned chlor-alkalai plant in Cartagena Bay was 1.7 ug THg g^{-1}, compared to a background level of 0.9 ug THg g^{-1} there during 2004-2005 (Olivero-Verbel et al. 2008). Yet, over a downstream distance of 3610 km, via the North Brazil and Loop WBCs (Walsh et al., 2011) through the Caribbean Sea and southern GOM, the mercury hair contents of Veracruz

residents were still 1.5 ug THg g^{-1} during 2005 (Guentzel et al., 2007) and 1.7 ug THg g^{-1} in 2011 (IPEN, 2013). Application of the aphorism "dilution is the solution to pollution" to paraphrase Garret Hardin's (1968) *Tragedy of the Commons* implies that other chlor-allai plants must have supplied additional mercury toxins en route, i.e. the Coatzacuakcis industrial area of Veracruz (IPEN, 2013), to offset physical mixing losses of this heavy metal poison.

Thus, the large values of THg (Shacklette et al., 1971; Rood et al. 1995) and p, p' DDT (Harner et al., 1999; Kannan et al., 2003), left within adjacent surface soils of the SE U.S., confirmed inferences of exported larger marine HABs as the causes of increased onshore fluxes in tongues (Fig. 2) of wind-borne marine aerosols. They were found after parallel trophic cascades and oil blowouts on both sides of the GOM impacted grazing pressures of the top-down controls (Walsh et al., 2015; 2016). But what lead to relaxation of the other bottom-up nutrient controls?

2.6 Oceanic Antillean boundary conditions at Puerto Rico and the Virgin Islands

Within a context of local sources of radionuclide tracers, not just more large-scale dispersion of bomb sources (Oktay et al., 2000), we now sort out local biotic processes from physically-controlled forcings at the land and oceanic boundaries, with analyses of radio-tracers of element cycling at upstream Puerto Rico and the downstream SAB. Rainfall over West Africa had declined significantly since 1900, with moisture of the soils there less during the last century, than over the preceding millennium (Hulme et al. 1994). Accordingly, greater frequencies of dust storms followed less rainfall in arid regions (Gillette, 1981), with larger amounts of dust asthma-triggers and iron-rich aerosols arriving in both North and South America, as well as in Europe (Fig. 4).

In response to an expanded Sahel, due to imprudent animal husbandry of overgrazing (Charney, 1975) and global warming (Held et al., 2005; Giannini et al., 2008) after 1970, the annual amount of Fe-replete mineral aerosols of Saharan Desert origin had increased 5-fold at 5000 km downstream of North Africa in Barbados, from 1965 to 1983 (Prospero and Nees, 1986). Once iron-replete *Trichodesmium* diazotrophs (Lenes et al., 2005) then fertilized HABs in coastal waters around Barbados, the prevalence of asthma among residents of Bridgetown had increased ten-fold, from 1.6% in 1970 (Pearson, 1973) to 19.5% by 2003 (Asher et al., 2006). During the intervening period of 1996, arrival of iron-rich dust at Barbados and pediatric asthma attendances at a hospital in Bridgetown during April of that year involved a time lag of one week (Prospero et al. 2008), which we attribute to **1**) Fe-fertilized growth of *Trichodesmium*, **2**) nutrient transfers to local HABs; and **3**) wind-borne delivery of their asthma triggers to these children.

Although earlier diazotrophs *Trichodesmium erythraeum* and *T. thiebautii* were also present at an even farther downstream distance of another ~925 km to the south Caribbean coast of Puerto Rico during 1958, HABs were rare there (Margalef, 1961), except for the eutrophic dinoflagellates *Pyrodinium bahamense* and *Cochlodinium polykrikoides* in small bays (Walsh et al., 2011). Accordingly, the fequency of asthma on Puerto Rico was then 5.4% in 1974 (Sifontes and Mayol, 1976), compared (Fig. 5a) to 30.3% in 1992 (Lara et al., 2006) and 41.3% by 2007 (Cohen et al., 2007), as one of the world's highest prevalence rate (Rodriguez-Cotto et al., 2013).

Concomitantly, the downstream population abundances of local HABs of *Ostreopsis lenticularis* at the bottom of the marine food web had increased 4-fold along Caracoles reef of SW Puerto Rico from 1983 to 1985 (Ballantine et al., 1988). Previously, as an index of other cuigateric HABs of *Gambierdiscus toxicus* in the same southwestern Puerto Rican region, some

of the top predator populations of *Syphreana barracuda* poisoned humans during 1980-1982 (de Motta et al., 1986). These fish predators then became more toxic there to humans, with a 6-fold increment of fall poisoning events by barracuda fish from 1985 to 1995 (Toteson, 2004), following maximal summer influxes of Saharan dust (Rodriguez-Cotto et al., 2013). These mineral aerosols fueled diazotrophs and eventually *Ostreopsis* spp., but not *Gambierdiscus*, which drew instead upon ubiquitous benthic interstitial nutrients of sediment pore waters.

Despite minimal nutrient- and light-limitations, the Caribbean HAB sources (McMillan et al., 1986) of ciguatoxins still declined by an order of magnitude both seasonally and interannually during 1983-1986 off southern Puerto Rico (Ballantine et al., 1988) and at Mattie Point in the British Virgin Islands during 1980-1981 (Tindall and Morton, 1998). By contrast, local increments of epiphytic *Ostreopsis* spp. were observed within both regions, i.e. in response to documented 5-fold increases of Fe-rich dust on Barbados, from 1965 to 1983 (Prospero and Nees, 1986).

The reputed toxicity attributed to colonial *Trichodesmium*, found later off St. Thomas in April 1990 (Hawser and Codd, 1992), like palytoxins described for this diazotroph in the South Pacific (Kerbrat et al., 2010), may actually have been due to resident *Ostreopsis* commensals, living among the colony's trichomes. On the Brazil shelf, *Ostreopsis* spp. similarly perch on the tops of floating *Sargassum* (Nascimento et al., 2006; 2012), while they are members of the drift communites off Florida (Bomber et al., 1988).

As an additional source of confusion, since both *Ostreopsis* and *Gambierdiscus* co-occur in ciguatera areas, it has been unwisely assumed that the former is also a source of fish poisoning to humans. But, no palytoxins occur in scarid vectors of at least blue humphead parottfish *Scarus*

ovifrons (Suzuki et al., 2013), while no known aerosols of ciguatoxins have been found, unlike those of *K. brevis* (Cheng et al., 2005a,b) and *O. ovata* (Ciminiello, 2014).

Thus, separate niches for all three HABs may be preserved within local coastal biophysical distillation processes [CBDP], effected equally by physical factors. As a reflection of habitat loss

from prior bombings of their nearby coral substrates around the Vieques archipelago, the populations of *Gambierdiscucs* along the south shore of Puerto Rico decreased ten-fold between 1982 and 1983 (Ballantine et al., 1988). Yet, recall that by 1995, Puerto Rican ciguatera events had returned (Toteson, 2004).

Furthermore, bottom cover of hard corals, *Orbicella annularis* = *Montastraea annularis,* along the 8-m nearshore isobath off St. John also declined from 45% there in 1987 to 7% in 2011 (Edmunds, 2013). More thermal bleachings occurred there (Manzello et al., 2007; Rogers et al., 2009), as also observed in Puerto Rico (Fig. 5a), in addition to Saharan sources of air-borne, deleterious soil fungi, *Aspergillis sydowii* (Shinn et al., 2000). Over the same time period of 1971-2011, the frequency of ciguatera events on St. Thomas had declined by 69% as well, from midpoints of 1975 to 2009, based upon presentations of human ciguatera poisonings in the emergency department of a local hospital (Radke et al., 2013).

Accordingly, we hypothesize that in the eastern Caribbean Sea, CBDP vectors of ciguatoxins returned, but not in the western sector, as a function of island size, and thus habitat heterogeneity. Consequently, we suggest that the original ciguatoxins were carried ashore to the larger land mass of ~13,800 km^2 for Puerto Rico by other HAB aerosol carriers – in some CBDPs, phthalates would be functional endocrine disruptor surrogates of DDT. Together with

multiple organic neurotoxins, the ciguatoxins would have been stored in land, freshwater, and marine reservoirs, as part of that Island's local hyrological cycles.

Whereas, a total land area of the American Virgin Islands of just ~345 km^2 may have been too small to accumulate adjacent marine environmental nearshore pools of multiple ciguatoxins, mercury poisons, endocrine disruptors, and asthma triggers in a CBDP. Indeeed, the comparable prevalence of asthma on Puerto Rico during 2002 was 11.5% , in contrast to 4.7% within the Virgin Islands that year (Rhodes et al., 2004). Of course, similar influxes of African dust pass Puerto Rico to arrive each summer above the WFS at an incremental downstream distance of another ~1975 km to impact fall plankton food webs and nearby humans (Lenes et al., 2001; 2012), followed by more local CBPD cycles there and elsewhere, en route to polar regions of North America.

Saharan dusts reach South America, as well. But, Brazil waters represented somewhat of a null case, in which *Karenia* and *Ostreopsis* carriers were present to cause asthma episodes and sea urchin mortalties. They also carried ashore, via wind dispersions, the co-travelling mercury poisons and endocrine disruptors of legacy DDT and phthalate esters, synthesized and applied over the last century. Finally, radio-iodine stimuli of thyroid cancer tracers there confirmed HAB deliveries of marine neurotoxins. Yet, coral reef bleachings alone were evidently too infrequent physical disturbances (Fig. 5a) of sufficient intensities to cause more than rare appearences of *Gambierdiscucs* on that shelf, with very few reported cases of ciguatera (Nascimento et al., 2012) in an otherwise typical tropical marine ecosystem.

2.7 Coastal biophysical pollutant distillation [CBPD] processes

Like relatively rare colonial breathing maladies off Veracruz in 1876, today's consequences of HAB asthma triggers from both *Karenia* and *Ostreopsi*s spp., as well as their

potential role in aerosol dispersion of ciguatoxins from *Gambierdiscus* spp., had escalated from just a few GOM and Caribbean observations of these food and pulmonary poisonings, before 1786 (Parra, 1787; Nunez Ortega, 1879). Once these recent HAB carriers were aloft over Puerto Rico, endocrine disruptions also prevailed ~two hundred years later. In 1969-1998 among young girls, phthalate esters of 1,2-benzenedicarboxylic acid occurred in their blood, together with premature breast developments, i.e. therlarche (Colon et al., 2000). Furthermore, rates of thyroid cancer in Puerto Rico were greater than those usually present in most of the mainland United States during 2007-2011 (O'Neil et al., 2015), except for U.S. coastal Counties situated along the SAB.

These phthalate esters were associated with chlorinated plasticizers and pesticides, as well as undescended testicles in boys, i.e. cryptorchidism (Thonneau et al., 2003). Phthalates are lipophillic and can be inhaled by humans (Autian, 1973; Cantonwine et al., 2014), but they are not natural products (Giam et al., 1978), no more than pesticides, nor long-lived radioiodine ^{129}I. First used as a plasticizer in 1920 (Viera et al., 2011), phthalate esters are now dispersed as another index of endocrine disruptions, like other legacy DDT/DDE compounds. Together with unexpected HAB carriers of MeHg aerosols, they have all spread from marine reservoirs through the terrestrial environment and humans, traced in some areas by radio-iodine effluents from nuclear: waste recycling facilities; power plants; and bomb tests.

Upon formation of these industrial phthalates, they inhibited growth of at least *K. brevis* (Wilson et al., 1978; Liu et 2016), and perhaps *Ostreopsis* spp. as well. Yet, laboratory surrogates of both naked dinoflagellates, *Gymnodinium simplex*, and of armored dinoflagellates, *Prorocentrum minimum* as representatives of the pelagic phases of *Karenia* and *Osteropsis,* survived as much as two-fold longer than diatoms, *Skeletonema costatum* and *Thalassioira*

pseudonana, at intermediate amounts of phthalate esters, contained within rubber tubings (Price et al., 1986).

Presumably, the greater toxicity of phthalates to herbivores *Acartia clausii* in the same experimental results of no surviving copepods (Price et al., 1986), together with different responses between diatom and dinoflagellate taxa, accelerated the latter HAB air-borne transfers of these additional organic pollutants to adjacent humans of Puerto Rico. In field studies off Piran, Slovenia selective seasonal feedings on *O. ovata* were exerted by both *A. clausii* and *Centropages* spp. (Furlan et al., 2013), but probably minimal toxin transfers to humans up through contaminated food webs occurred via the copepod vectors of the water column, since this other ungrazed tracer of human endocrine disruptions – besides pesticides – survived to accumulate on sea bottoms and within the overlying air parcels.

Indeed, the most abundant fat-soluble phthalates, i.e. high molelcular weight di(2-ethylhexyl)phthalate [DEHP] during 1999 in the Dutch Wadden See exhibited the usual organic pollutant pathways of a sedimentary sink, with minimal degredation in water allowing some pelagic trophic exchanges, and a net export to air, due to physical processes of sedimentation and vapor pressure (Peijnenburg and Struijs, 2006). In the sediments, *O. ovata* would have been consumed by mussel herbivores (Ceredi et al., 2010; Amzil et al., 2012; Fraga et al., 2012; Ciminiello et al., 2015), as one of the benthic routes of palytoxin and phthalate poisonings to sea urchin and human predators.

Furthermore, phytophagous fish of coral reefs, e.g. parrotfishes, not anchovy herbivores of the overling water column, would instead have served as another ciguatera vector, after those reef herbivores ate attached algae and perhaps both of the very toxic epibenthic HABs in shorter food chains. Direct measurements of ciguatoxins and maitotoxins of *Gambierdiscus* spp. origins

(Lewis et al., 2016) have been found in tissues of parrotfish *Scarus gibbus* (Satake et al., 1997) and *S. russelii* herbivores – as designated by $^{15/14}$N ratios (Mak et al., 2013). Yet, in other parrotfish *S. ovifrons*, no palytoxins of *Ostreopsis* spp. origins were found thus far (Taniyama et al., 2003; Suzuki et al., 2012; 2013).

Akin to other anthropogenic perturbations of coral reef substrates during construction of military bases (Ruff, 1989), persistent practice bombing of Vieques Island by the U.S. Navy over ~60 years had little (Antonius and Weiner, 1982), or major (Porter et al., 2011; Hernandez-Delgado et al., 2014), impacts on the nearby coal reefs. Yet, they harbored *Ostreopsis* HAB (Ballantine et al., 1988) carriers of marine aerosolized (Ciminiello et al., 2014) organic toxins, presumably including MeHg, DDT/DDE, and again unexpected ciguatoxins. Physical disturbances, such as temperature-caused coral reef bleachings (Fig. 5a) and/or wayward bombs, would have encouraged both of these tychopelagic and benthic, very toxic dinoflagellates to leave their coral substrates and enter the overlying water column, placing them closer to adjacent human victims. At times, other HABs of *Gambierdiscus toxicus* can be free-swimming as well (Parsons et al., 2011).

Consequently, despite the near-field release of mercury via the Rio Grande de Loiza River to the northeast of San Juan from the chlor-alkalai plant at Caguas, the hair content of mercury among residents of Vieques Island off the farther-field east coast during 2000-2002 was a large amount of 9 ug THg g^{-1} (Ortiz-Roque and Lopez-Rivera, 2004). In comparison, less efficient onshore transfers by unbombed, wind-blown HABs to people living in NE Puerto Rico resulted in nine-fold smaller heavy metal contaminations, as monitored in human hair of just 1 ug THg g^{-1} (Ortiz-Roque and Lopez-Rivera, 2004), when not carried ashore by physically-disturbed tychopelagic dinoflagellate epiphytes of coral reefs.

Finally, this Islands's radio-iodine tracers were part of the ~530 megabecquerels [MBq] of radio-activity emitted to the atmosphere during 1965-1968 from the poorly designed Boiling Nucelar Superheated [BONUS] reactor at Rincon, Puerto Rico and ~17 MBq released during ecological experiments within the El Verde tropical forest over the same time period (Odum and Pigeon, 1970). Yet, by contrast, these were still very small amounts of Puerto Rican radioactivity – much less than the amount of the radiation evaded to the troposphere during the Chernobyl incident.

To the extent that Puerto Rico's local hydrological budget of water is in balance, annual rainfalls may approximate those of freshwater runoffs in a second tropical CBDP. With the assumption that human water diversions for agriculture and drinking over Puerto Rico do not yet exceed wet depositions, sewage sludge pools of phthalate esters would also eventually reach the surrounding seas, e.g. within the longest urban river of Rio La Plata in northeastern Puerto Rico during the rainy season (Ortiz-Colon et al., 2016).

In coastal waters, evaporation and onshore influxes of moisture complete the water cycle. While in the same places and times, malign marine HAB DOC-modulated aerosols of brevetoxins and palytoxins, fueled by no-longer Fe-deficient resident nitrogen-fixers, would import both far-field MeHg and DDT/DDE (Garrison et al., 2014) neurotoxins. They would also return the local Puerto Rican phthalate pollutants, exported from the Island by river and ground waters, as part of a repeated CBPD aggregation of concentrated persistent organic pollutants [POPs]. Where physical disturbances, i.e. bombing of the Vieques coral reefs, suspended ciguatoxins, they would would become air-borne as well, co-carried by *Ostreopsis* and *Karenia* aerosols. Yet, equipotent maitotoxins of *Gambierdiscus* spp. origins are water soluble, not

lipophilic like ciguatoxins, so less is known about their roles in fish poisonings (Van Egmond et al., 2004; Lewis et al., 2016).

2.9 Riverine boundary conditions in the SAB

"Anomalous" high rates of thyroid cancer were reported among residents of Charleston County, South Carolina, during 1996-2000, i.e. greater than expected, compared to the rest of the state (SC Central Cancer Registry, 2003). During 1996, the State of South Carolina had also issued fishing advisories, based on concerns about both the mercury and radionuclide levels within the Savannah River (Burger, 1998). Moreover, in hindsight, the most likely regional source of radio-iodine was the Savannah River Laboratory [SRL], which housed five nuclear reactors and additional waste management facilities thoughout the 1950s-1980s.

In a subsequent survey of North American rivers, using sensitive accelerator mass spectrometry techniques, the concentrations of ^{129}I of a long half-life of 15.6 million years were measured in the Savannah River, flowing into the downstream SAB during 1994. These dissolved isotope stocks were then ~three orders of magnitude greater than those of the individual Appalachicola, Mobile, Mississippi, and Rio Grande Rivers draining into the GOM (Moran et al., 2002; Kaplan et al., 2011).

Given the persistence of the radioactive decay of ^{129}I, after closure of the upstream SRL nuclear reactors by 1992, the rates of thyroid cancer during 2005-2006 were still anomalously high (Mangano, 2011). Indeed, over six years, thyroid carcinomas had increased 74% from 1999-2000, within the metropolitan statistical area of the surrounding South Carolina and Georgia Counties, compared to a 31% increment for all of the U.S., over the same time period (Mangano, 2011).

Thus, at the mouth of the Savannah River in the SAB, both HABs there of upstream WFS origins (Murphy et al., 1975; Tester et al., 1991; Steidinger et al., 2008; Walsh et al., 2009) and radio-iodine of SRS origins, together with other local halides, would have awaited downstream and onshore co-transports during storms. In the SAB, sea spray was routinely deposited inland about 400 km from the coast (Junge and Gustafson, 1957), at a similar local rate of sea salt fallout as occured along the west coast of Florida (Brierly, 1965). Moreover, we shall find that the cumulative releases of ^{129}I from the SRL had been only 1-2% of the radioactive effluents of this same long-lived isotope from the European nuclear waste reprocessing plants at Sellafield, England and LaHague, France (Snyder et al., 2010).

Furthermore, after transit along the intervening Georgia shelf, the hair mercury contents of residents within the Charleston environs was a mean (Table 2) of 2.7 ug THg g^{-1} by 2007 (Bartelme, 2007). The presumed sources of this heavy metal contamination were at least two regional major upstream primitive Castner-Kellner chlor-alkali plants, using a mercury technique - developed in the 1890s - for the electrolytic productions of caustic soda and chlorine gas.

These known Georgia Hg supplies to the SAB were located in Augusta, next to the Savannah River as an operational source of mercury pollution for ~45 years over 1966-2010 (Smith et al., 2007), and in Brunswick, along the Atlantic shoreline as an EPA superfund site of mercury contamination, after another ~40 years of Hg releases during 1955-1994 (Kannan et al., 1997). By 1973, deleterions impacts of mercury poisoning had occurred there in local fishes and avifauna (Odom, 1975). Significant amounts of methyl mercury were also found in both gastropod *Littorina irrorata* and fiddler crab *Uca pugnax* detritivores (Windom et al., 1976). In 1989, mercury within sediments of the recipient Turtle River estuary were still very toxic to amphipod herbivores, *Hyella azteca* (Winger et al., 1993). Finally, consumption advisories about

which fish herbivores and zooplanktivores to not eat more than once each month, from impacted surrounding areas of Glynn County, were still in effect during 2016.

Another subsequent EPA superfund site of mercury neurotoxin discharge also involved heavy metal exports during 1966-1979 from Lavaca Bay, Texas (Sager, 2002; Bloom et al., 2004). These Hg inputs contributed partly to the same hair mercury content found in Alabama of 2.8 ug THg g^{-1} (Warner, 2007). Thus, via upstream WBCs of the Loop Current and the Gulf Stream, as well as due to migratory movements of adult barracuda, vectors of methyl mercury and ciguatera poisonings moved to Georgia and South Carolina

Furthermore, over the same time, the prevalence of asthma among children in Charleston had increased ~14-fold from steady low values during 1958-1972 to many more malignant breathing episodes by 1997 (Crater et al., 2001). These pulmonary maldies reflected the known imports of pelagic *Karenia* spp. HAB carriers to the GOM, past Georgia to North Carolina, as cited above. By 1964-1968, when greater asthma indices of HAB wind-borne, malignant marine pulmonary trggers were beginning to change in Charleston, putitive HABs of *Gymnodinium* spp. were becoming more frequent in the SAB (Marshall, 1971).

Upstream at the southern end of the SAB within the Florida Keys, range extensions of the dominant pelagic HABs of *Karenia* spp. and the more potent tychopelagic HABs of *Ostreopsis* and *Gambierdiscus* spp. had also occurred from 1979 to 2009 (Taylor, 1979; Norris et al., 1985; Okodlokov et al,, 2007; Villareal et al., 2007; Litaker et al., 2009; Parsons et al., 2012; Walsh et al., 2015), with subsequent ciguatera poisonings reported as far north as in Duval County during 2000-2011 (Radke et al., 2015). Thus, it was not surprising that in August 2004, a ciguatera event had also occurred from people eating adult barracuda, caught ~100 km southeast of

Charleston (Villareal et al., 2006), during alongshelf migrations of these piscivore predators (O'Toole et al., 2011; Hansen and Kerstetter, 2015).

Although *Trichodesmium* and other diazotroph sources of HAB nutrient supplies were evidently present along the Georgia coast at St. Catherines since 1075, based on an isotope budget (Walsh et al., 2009) and $^{14/15}$N ratios of exhumed earlier human residents (Schoeninger et., 1983), the abundances of at least the colonial nitrogen-fixers recently increased there during 1974-1998 (Dunstan and Hosford, 1977; Subramanian et al., 2002). After 5–fold upstream increments of air-borne, iron-rich Saharan dust loadings at Barbados during 1965-1983 (Prospero and Nees, 1986), when *Tichodesmium* colonies amounted to 71-76% of the total phytoplankton commuities near the Gulf Stream western boundary current of the SAB off Georgia, their chlorophyll biomasses summed to 1.8 ug chl l^{-1} in 1974 (Dunstan and Hosford, 1977), compared to 3.1 ug chl l^{-1} in 1998 (Subramanian et al., 2002). Concomittantly, one potential HAB, *Gymnodinium* spp = possibly *Karenia* spp. was seasonally dominant earlier on the SAB shelf during 13 cruises in 1964-1968 (Marshall et al., 1971).

Finally, by 1983 at Hollywood, South Carolina within the drainge basin of the Edisto River in Charleston County, concerns about accumulations during 1969-1974 of more POPs, including phthalates, in another CBDP led to a designation there of an additional EPA superfund site. Similarly, when this diazotroph reprised its SAB role (Hutchinson, 1965) as one of the casual agents of HAB expansions in other regions, two of these commensal HABs permeated the western boundary Brazil Current off South America as well. In the absence of many *Gambierdisus* spp., wind-driven *Karenia* and *Ostreopsis* spp. evidently still carried ashore there combined DOC triggers of POP-causing asthma, endocrine disruption, hydragyria, and thyroid cancer episodes.

3.0 South American analog

With a much greater land area of ~8, 514, 200 km^2, Brazil is the third case of a tropical CBDP, albeit a large one, with anthropogenic alterations of both its hydrologicial cycle and nutrient loading to coastal seas. Reduced evapotranspiration of the diminished Amazon rainforest resulted in less wind-borne regional freshwater supplies within *flying rivers* (Marengo et al., 2011; Nobre, 2014), from the most deforestated State [23°90'S, 62°76'W] of Rondonia to yield smaller downstream rainfalls in Sao Paulo. The resultant recent draughts there during 2014-2016 were the worst in 80 years, reminiscent of a *once-in-century drought of 2005* in Brazil (Marengo et al., 2011).

To compound human asthma breathing problems in Sao Paulo, as well as thyroid cancers and water shortages, in addition to mercury, phthalate, and DDT poisonings there, smoke from burning rainforest fires followed other aerial trajectories south from the Amazon River drainage basin. However, the same hoary Navier-Stokes equations of the 19th century can be applied to both air (Sanchez-Ccoyllo et al., 2006; Adhikary et al., 2007; Mamun et al., 2014) and water fluid motions of different densities, once obtuse validation data sets - in white pebble Hansel and Gretel fashion - provide hints about the underlying, non-linear interactions of pollutant perturbations.

For example, tropical deforestations also yielded increments of estuarine nutrient loadings from replacement pasture soils and leachates, with smaller N/P molar ratios of ~2, compared to previous forested watershed nutrient export ratios of ~16 (Buschbacher, 1986; Downing et al., 1999; Neil et al., 2001; Valiela et al., 2013). Thus, growth of all nitrogen-fixers were favored within nearshore recipient marine waters. After intensive deforestation of Brazil began in small farms (Fearnside, 2005) during the 1960s, the N/P ratio of the Amazon and

Tocantins River effluents was ~5 by the 1980s (Howarth et al., 1996). By contrast, the N/P ratio was then 38 in the Mississippi River (Howarth et al., 1996).

3.1 Nutrient constraints, phytoplankton responses, and reduced herbivory

Near the mouth of the Amazon River at Belem [1°27' S], phosphorus-replete N/P ratios previously allowed both *Trichodesmium* and diatom diazotroph associations [DDA] to flourish, until a combination of nitrogen- and silica-limitation ensued, as observed within the Mississippi River Delta (Dortch and Whitledge, 1992). The nearsurface molar NO_3/PO_4 and SiO_4/NO_3 ratios of dissolved nutrients within the ~30-m euphotic zone at mid-shelf above the 45-m isobath had been respectively <1 of a phosphorus-replete and >5 of a silicon-replete Brazilian chemical habitat during May-June 1976 (Edmond et al., 1981)

These nutrient stocks on the inner NE Brazil shelf were similar to observations obtained a decade earlier (Ryther et al., 1967) and had already *set the plankton stage* (Hutchinson, 1965) for estuarine diatoms, harboring endosymbiont nitrogen-fixers. They included *Richelia intracellularis* within DDAs there in 1996 (Carpenter et al., 1999) and 2001 (Shipe et al., 2007; Subramanian et al., 2008), as also found off the Mekong River Delta (Grosse et al., 2010).

Whereas, at the seaward edge of both these tropical Brazilian and Vietnamese shelves, the oceanic colonial marine diazotrophs *Trichodesmium* spp. were the major nitrogen-fixers under less silicate stocks. Indeed, farther offshore, based on these and other silicate data sets (Milliman and Boyle, 1975; Froelich et al., 1978), continued diatom biotic removal of silica from the far-field Amazon River plume would have been unlikely, compared to conservative physical mixing losses. Even on the Amazon shelf there was very poor correlation between phytoplankton

particulate pools of biogenic silicon and organic carbon during 1989-1991 (DeMaster et al., 1996), due to abundances of other non-siliceous primary producers, i.e. those of HABs.

Furthermore, earlier respiratory and coughing problems of "*Tamandare fever*" among humans living near Recife in Pernambuco State, Brazil [8°03'S] had been mistakenly attributed to just *Trichodesmium* itself during the 1960s (Sato et al., 1963; Proenca et al., 2009). Nevertheless, more recent co-occurrences of both *Trichodesmium* and *Ostreopsis* spp. were instead found a half century later along the Brazil coastline (Penna et al., 2005; Nascimento et al., 2006; Graneli et al., 2011), where asthma prevalences of 19.1-20.1% among schoolchildren occurred in 1994 and 2003 (Mallol et al., 2000; de Britto et al., 2004; Sole et al., 2006). These asthma attacks had since increased by 2006 (Dela Bianca et al., 2010) to 26.6% in Sao Paulo [23°33' S], where local marine waters were instead dominated by *K. mikimotoi* and *Trichodesmium* spp., unlike off Chile (Jackson et al., 2011) within the diatom-rich coastal upelling WBC ecosystem of the Humboldt [Peru-Chile] Current (Table 2).

In Rio de Janeiro State, the dissolved N/P ratio of Brazilian seawater during 1980-1993 at the mouth of Guanabara Bay [22°50'S] was also just 9.3, more typical of sewage effluents and surrounded by the cities of Rio de Janeiro and Niteroi (Ribiero and Kjerfve, 2002). Associated with the local population increases of humans and their excretory sources of phosphorus, the P influxes to sediments of Guanabara Bay had increased nine-fold over the last century, since 1875 (Borges et al., 2009). These phosphorus enhancements of South American coastal waters would have continued to favor nitrogen-fixers, of course, as noted earlier by Charles Darwin's observations of *Trichodesmium* spp. there during Saharan Desert dustfalls of the 19th century.

Farther south on the inner shelf of Parana State [25°42' S], a smaller nitrogen-depleted NO_3/PO_4 ratio of only 0.8 was also found in surface waters (Fernandes and Brandini, 2004), i.e.

without consideration of ubiquitous di-nitrogen gas, as a consequence instead of faster P recycling of nearshore decomposing diatom populations (Walsh et al., 2006). Off Brazil, siliceous net phytoplankton of >20 microns sizes occurred during both seasonal upwelling periods (Guenther et al., 2008; Kutter et al., 2014) and at the mouths of the Amazon, Sao Franciso, and La Plata Rivers (Brandini et al., 2007; de Rezende et al., 2015; Medeiros et al., 2016).

Accordingly, off Parana State *Trichodesmium* accumulated there as much as 65 µg chl l^{-1} of these colonial diazotrophs (Siqueira et al., 2006). Concomitantly, remote satellite-sensed signals of these diazotrophs were most frequently observed at 25°S (Westberry and Siegel, 2006). Other *Trichodesmium* blooms washed ashore on Uruguayan beaches at 35°S (Mendez and Medina, 2004), and *vast swarms* of *Trichodesmium* were observed, between Montevideo and South Georgia Island, at 40°S (Hart, 1934).

Earlier during 1978 and 1981, prior cryptic populations of *Karenia mikimotoi* = *Gyrodinium auroleum*, rather than *O. ovata*, had first emerged as HABs along Hermenegildo Beach [33°39'S] in the southernmost Brazilian State of Rio Grande do Sul (Proenca, 2006), i.e. after the expanded Sahel export of desert dust to the Americas began in 1965 (Prospero and Nees, 1986). Off Brazil, both fishkills and human breathing difficulties accompanied strong waves during formation of aerosolized gymnocin toxins within sea spray, when these HABs also occuured in both years off Brazil at ~32°S (Rosa and Buselato, 1981; Tomasi, 1983).

Farther equatorwards along the southeast coast of Brazil, >70 subsequent oil spills in just Sao Paulo State during 1980-1990 (Heilemann and Gasalla, 2008) had decimated zooplankton herbivores (Table 1) to set free local HABs. Off Rio de Janeiro, the more toxic tychopelagic HABs of *Ostreopsis ovata* and prior cryptic *Gambierdiscus toxicus* were noted,

with their epiphytic niches observed on at least floating *Sargassum* (Nascimento et al., 2012a; b). These latter phytoplankton sources of ciguatoxins had finally emerged, after fewer coral reef bleachings in Brazil (Costa et al., 2000), compared to Puerto Rico (Fig. 5a).

Concurrently by 1988, a ~60-km wide HAB of as much as 16 ug chl l^{-1} of *K. mikimotoi* was found on the Argentine outer shelf, above the 85-m isobath at 39°15'S (Negri et al., 1992), i.e. HAB carriers of POPs, heavy metals, and radionuclides. Moreover, upon autopsies, occult papillary thyroid cancers in Argentina were ~two-fold more frequent than those in Portugal (Ottino et al., 1989), where diatoms were instead the dominant phytoplankton of that EBC (Table 2). Moreover, hair mercury contents of Portuguese sea-side residents, next to the diatom-rich EBC of the seasonal Portuguese Current, were also 5-fold less than people, impacted by the comparable WBCs of the Gulf Stream off England (Table 2) and the Brazil Current off South America.

3.3 Mercury loadings

Where artisanal, Hg-mediated gold-mining occurred along the banks of the Tapajos River tributary of the Amazon River, the hair content of total mercury [THg] in Brazilian miners was ~29.5 ug THg g^{-1} during 1992-1994 (Malm et al., 1995; Akagi et al., 1995; Barbosa et al., 1997), compared to 3.0 ug THg g^{-1} at Belem in 2004 (Pinheiro et al., 2006), i.e. a ten-fold decline near the Amazon Delta among an unknown number of miners. They had all inhaled mercury aerosols, using ancient methods to extract either gold and/or silver. During 384-287 BC, both Aristotle and Theophrastus described removals of the heavy metal mercury from cinnabar ore, also known as both vermilion and mercuric sulfide [HgS], to be used as orange-red pigments.

By simply heating the ore with moderate fires, evasion of the toxic Hg aerosol occurred. Next condensation of this gaseous stage led to liquid elemental mercury at room temperatures for extraction of gold and silver from their mineral deposits. After the gold-mercuy amalgam was formed, gold was extracted by again heating to evaporate the mercury, once more exposing the user to inhalation of this very toxic element. Accordingly, hair mercury of artisanal miners just tells one where the gold was harvested, while hair mercury of human residents next to chlor-alkali plants instead provides information about transfers of industrial pollutants to downstream victims, via a combination of environmental vectors and the amounts of initial Hg loadings.

Upstream next to coastal Recife, smaller heavy metal amounts of 1.9 ug THg g^{-1} were found in human hair, due to both inhalation and fish food within Itapissuma in 1999, adjacent to the more usual mercury source of a smaller chlor-alkali plant (Nilson et al., 2001). Poleward, along SE Brazil, residents of Rio de Janeiro, who did not eat fish, had a hair content of 1.7 ug THg g^{-1} during 1987 (Malm et al., 1990). In that same year, a larger hair amount of 5.4 ug THg g^{-1} occurred in fish-eaters (Malm et al., 1990), i.e. at least 26% of the hydragyria cases in Rio de Janeiro were then presumably due to inhalation.

Yet, the residents along the Santa Cruz channel and the Santos estuary had only 0.4 ug THg g^{-1} in their hair during 2007 within the Sao Paulo environs, despite eating fish (Farias et al., 2014). Thus, the two major commercial chlor-alkali facilities in upstream Todos dos Santos and Guanabara Bays of Brazil's largest brackish ecosystems dominated the water- and air-borne onshore fluxes of legacy mercury. Similarly, the latter Bay was also the source of a radio-tracer for analysis of other anthropogenic POP redistributions.

3.4 Endocrine disruptors

In terms of pesticide endocrine disruptors in human breast milk during 1988-1992 within southern coastal Brazil cities of Rio de Janeiro, Sao Paulo, and Porto Alegre, the mean DDE/DDT ratio was ~11.0 (Lara et al., 1982; Beretta and Dick, 1994; Paumgartten et al., 2000). Similarly, the DDE/DDT ratio was 14.3 during 1999 in Sante Fe, Argentina, at an inland distance of 395 km from coastal Buenos Aires (Munoz-de-Toro, et al. 2006). Finally, along the Madeira tributary of the Amazon River, human breast milk contained a ratio of ~4.8 during 2001-2002 (Azeredo et al., 2008). These data all suggest varying legacy applications of these pesticides to deal wth malaria, but their malign health impacts have now been replaced by other industrial neurotoxic phthalate POPs - as additional endocrine disruptors.

This concurrent index of CBDP pollutant transfers was studied within adjacent French Guiana. Since human activities are the sources of phthalate neurotoxins, an expected onshore gradient was noted between their occurrences in rainforests and sea-side cities during 2013 (Lenoir et al., 2016). Ten-fold larger amounts of these hormone disruptors were found in urban areas, with inferences of POP origins and inland aerosol transports (Lenoir et al., 2016). A similar seaward gradient of increasing dissolved phthalate concentrations was observed in Guanabara Bay (Loureiro et al., 2001), where estrogens also prevailed in sewage outfalls (Ternes et al., 1999).

Finally, large amounts of phthalates were found in surrounding soils of Sao Paulo, to form another facet of this Brazilian offshore riverine and onshore wind-blown CBDP. But, since local industrial factories (Ferreira and Morita, 2102) were also a source of this POP, additional data are required to construct a complete budget of endocrine disruptions there (Gimilani et al. 2016). However, unfortunately a radiotracer did provide unwanted information on the likely sign for the cross-shelf directions of all these CBDP environmental pollutant transfers off southern Brazil.

3.5 Radiotracers and air parcel back-trajectories

A radiotracer of these POP transfers, effected by the changing latitudinal composition of the wind-blown phytoplankton communities, between Brazilian coastal seas and adjacent humans, was also released there in the form of short-lived radio-iodines, $^{131\text{-}135}$I, from nuclear facilities in Guanabara Bay. Despite ephemeral half-lives, those radionuclides would have not all decayed. The remaining mutagens would still have induced thyroid carcinomas during the short alongshore, southward transits of near shore waters and sea sprays from Rio de Janeiro to Sao Paulo, before moving downstream to Argentine waters.

Consequently, the ill health impacts of these inferred downstream HAB transports of ionizing radiations from $^{131\text{-}135}$I were confirmed by observations of thyroid carcinomas among men and women along the Brazil coastline during 1996-1998 (Coeli et al., 2005; Veiga et al., 2013). In Sao Paulo [23^{o}33' S], thyroid cancer prevalences were ten-fold greater than those found among residents of Belem [1^{o}27' S], in the same years along the Para distributary of the Amazon River. At the shoreline of Itaorna Beach in Rio de Janeiro [23^{o}00' S, 44^{o}27' W], two pressurized water nuclear reactors at the Angra plant had released (Chanrasekaran et al., 1985) $^{131\text{-}135}$I of <60 day half-lives since 1985, without additional consideration of long-lived ^{129}I amounts (Hou et al., 2992), considered here in the last analogous coastal ecosystems of Oceania.

Under wind forcings from the northeast, such short-lived Brazilian iodine mutagen agents would have drifted south at water current speeds of as much as ~10 cm sec^{-1}, or ~8.6 km day^{-1}, towards Sao Paulo in a few weeks over the ~400-km alongshore distance from Rio de Janeiro. Over this short time period of ~47 days, the observed thyroid cancers could have been induced by some of the remaining undecayed iodine radio-isotopes, accrued within wind-blown onshore sea sprays and HAB carriers.

If the radio-iodine cancer tiggers became air-borne en route, as aerosols within wind-forced sea sprays, their arrival times among downstream human victims would have been much less, of course. At wind velocities of ~5 m sec^{-1}, or ~345 km day^{-1}, transit times of short-lived iodine radio-isotoes from Rio de Janeiro to Sao Paulo would instead been ~1.2 days, well within their radioactive decay periods. Upon arrival above the Sao Sebastio Channel [23°48'S] of Sao Paulo State as lysed radioactive DOC sources of wind-blown neurotoxins, their presence there would have been reflected in **1**) the increment of asthma attacks in Sao Paulo from 1994 to 2003 (Mallol et al., 2000; de Britto et al., 2004; Sole et al., 2006; Dela Bianca et al., 2010), **2**) presences of sea salts (Bourotte et al., 2011), and **3**) the concurrent ten-fold higher rates of thyroid cancer (Coeli et al., 2005; Veiga et al., 2013).

Even during seasonal winter reversals of the alongshore flows of the Brazil Current towards Uruguay and Argentina (Lima et al., 1996), 4-day back-trajectories from an atmospheric circulation model of those air parcels, which arrived above the sea shores of Sao Paulo and adjacent Cunha, Brazil, indicated that 32% came from above the NE Atlantic Ocean (Sanchez-Ccoyllo et al., 2006). By contrast, another 12% of these trajectories travelled above SE shelf waters (Sanchez-Ccoyllo et al., 2006), such that a total 44% of the arriving winter aerosols may have been entrained marine HABs within sea sprays from coastal waters, as opposed to terrestrial aerosols of *flying rivers* from Brazil's interior.

Moreover, 57% of these putative marine aerosols then had diameters of <10 microns as measured at Sao Paulo shore stations (Sanchez-Ccoyllo et al., 2006), similar to aerosol sizes of 5-6 microns for *K. brevis* (Cheng et al., 2005) and *O. ovata* (Cinminiello et al., 2014). Finally, the Na/Cl content of aerosols at Cunha was typical of sea salts at that time (Bourotte et al., 2011). Thus, the lysed DOC asthma triggers of these dinoflagellate HAB carriers of *Ostreopsis* and

Karenia spp. HABs, fueled by diaotrophs and entrained within sea salts, would have carried ashore both organic mercury and pesticide poisons, as well as radionuclides and other endocrine disruptors, during their onshore, wind-borne transports to human victims within sea sprays off Sao Paulo. These same processes led to the same results in other analogs of European, Indian, African, Asian, and Oceania waters.

4.0 European analog

Europe is also downwind of the Saharan Desert, such that infrequent dust storm exports to England and Ireland during 1903-1968 had become almost monthly events during 1991 (Goudie and Middleton, 1992). Subsequent blooms of colonial *Trichodesmium* spp. appeared within the English Channel by 2003 (Fig. 6), compared to infrequent occurrences of these diazotrophs within Irish coastal waters during the 1930s (Farran, 1932). After diazotrophs fueled dinoflagellate HABs in European waters, and oil spills (Fig. 5b) decimated copepod herbivores there (Table 1), asthma prevalence among adjacent humans increased exponentially from 1950 to 2000 (Fig. 5b).

4.1 Signatures of multiple nitrogen-fixers

At seasonal low temperatures of <20^{o}C within the English Channel, however, growth of *Trichodesmium* spp. there would have sufferered thermal constraints, as well as seasonal Fe- and P-limitations (Breitbarth et al., 2008; Fernandez et al., 2010). By 2006, unicellular, presumably free-living diazotrophs were also found in the Channel water column, as expressed by multiple nif genes (Rees et al., 2009). Yet, endosymbiont nitrogen-fixers, e.g. *Richelia intracellularis* within DDA, also tended to occur in subtropical and tropical regions (Carpenter et al., 1999; Bar Zeev et al., 2008; Grosse et al., 2010).

During continued NW penetrations of Saharan dust plumes (Franzen et al., 1995) above

the English Channel, including March 1998, dissolved iron concentrations of surface seawaters near Plymouth, England, increased then to ~4.0 nmol Fe l^{-1}, compared to background values of 0.5 nmol Fe l^{-1} at the shelf-beak (deJong et al., 2007). After seasonal arrivals of Saharan dust above the outer parts of the WFS, dissolved Fe stocks similarly increased ten-fold there, from 0.3 to 3.0 nmol Fe l^{-1} during 1998 and 1999 (Walsh et al., 2006).

Accordingly, with large half-saturation constants of ~0.5 nmol Fe l^{-1} for dissolved iron uptake by *Trichodesmium*, compared to 10-100 fold smaller values for iron assimilation by bacteria, diatoms, and dinoflagellates (Walsh et al., 2015), enhanced specific growths of diazotrophs would still have prevailed within these episodic, Fe-rich chemical habitats. Presumably, their P-limitation in the English Channel environs was also minimal during either summer nutrient recycling events (Cooper, 1933; Armstrong et al., 1974; Pingree et al., 1977; Laane et al., 1993; Jordan and Joint, 1998), or near sewage outfalls.

At higher trophic levels, greater marine nitrogen-fixation within the North Sea was also apparent in another time series of consumer body isotopes, i.e. harbor porpoises *Phocoena phocoena* (Christensen and Richardson, 2008). Their $^{14}N/^{15}N$ ratios displayed the isotopic signatures of diazotrophs, after passage though consumers (Walsh et al., 2009). These data suggested that the contribution of dinitrogen gas to primary production experienced a multi-decadal increment in phosphorus-replete summer waters of <6 N/P ratios within the English Channel (Jordan and Joint, 1998). Off the Dutch coast of the North Sea, the inferred nitrogen-fixation had increased ~10% from 1848 to 2002 (Christensen and Richardson, 2008) in the second long-term nitrogen isotope time series of this review. Another one will be of mercury in wigs of human hair in Japan (Suzuki et al., 1984), while the last one is comprised of ^{129}I concentrations within Indo-Pacfic corals (Biddulph et al., 2006).

4.2 Removal of top-down controls

Like the increased human asthma attacks after bleached coral reefs of tropical and subtropical regions (Fig. 5a), an exponential increment of asthma episodes among western European school children had also occurred between 1953 and 2003, after major oil spills (Fig. 5b). Concurrent overfishing, radioactive waste exports, pesticide applications, phthalate dispersals, and mercury releases from 47 sea-side chlor-alkalai plants in Europe prevailed by 2001. But, evidently, it was mainly the repeated oil spills of the *Torrey Canyon*, *Tsesis*, *Amoco Cadiz*, *Betelguese*, *Braer*, *Sea Empress*, and *Prestige* tankers in the English Channel environs that first killed copepods (Table 1), setting free diazotroph-fueled HABs to instead be lysed by bacteria as asthma triggers. The brief *Ekofisk Bravo* blowout of ~3.21 x 10^4 m^3 crude oil also occurred in Norwegian waters during 22-30 April 1977.

First recorded in European waters off Norway during the autumn season of 1966 (Braarud and Heimdal, 1970), the HAB stage of these prior cryptic phases of *K. mikimotoi* had later emerged off Plymouth, England, after March 1967 (Boalch, 1987). Then, oil spills of the *Torrey Canyon* initially led to ten-fold reductions of copepod herbivores within the same water masses of the English Channel (Southward et al., 1995). There, subsequently both *Trichodesmiuum* precursors, dating from at least 1910 in the Irish Sea (Hjort, 1911), and HABs of *K. mikimotoi* became annual events (Figs. 6a, b), because top-down perturbations of the food web continued.

Additionally, other declines (Southward et al., 1995) of oiled copepod grazers *Calanus helgolandicus* occurred off both Plymouth, England (another vertical red line of Fig. 5b) and along the French Atlantic coast (Laubier, 1980; Samian et al., 1980), after this second set of large [>1 x 10^5 m^3 crude oil] *Amoco Cadiz* spills was released during March 1978 in the English

Channel. A similar decrement of local populations of other herbivores *Paracalanus* and *Pseudocalanus* spp. was also observed during 1978 in the German Bight off Helgoland (Greve et al., 1996). There, *K. mikimotoi* had first emerged in August 1968 (Hickel et al., 1971), after the *Torrey Canyon* spills.

4.3 Inferred pulmonary consequences

On the Ilse of Wight in the middle of the English Channel, the frequencies of total childhood asthma cases (Fig. 5b) increased from 2.3% in 1964 (Graham et al., 1967), before oil spills of the *Torrey Canyon*, to 40.3% prevalence of asthma afterwards in 1999 (Tariq et al., 1998; Kurukulaaratchy et al., 2003; Arshad et al., 2005). Within nearby seaside Copenhagen, Denmark, the prevalences of asthma increased from a background value of 0.8% in 1957 (Frandsen, 1958) to 5.3% in 1986 and 16.0% by 1991 (Backer and Ulrik, 1992), after these initial petrochemical perturbations left behind ungrazed HABs.

The subsequent spills of crude oil from the supertanker *Braer* off northeastern Scotland in the North Sea during 1993 (Fig. 5b) amounted to 9.4 x 10^4 m^3 (Campbell et al., 1993), similar to the *Torrey Canyon* spills of 1.4 x 10^5 m^3. The frequency of asthma among children in nearby Aberdeen, Scotland at the coast increased from 4.1% in 1964, before the *Braer* spill, to 20.0% afterwards in 1994 (Ninan and Russell, 1992; Devenny et al., 2004).

We attribute these temporal changes of Scottish asthma prevalence among school children to altered, oil-poisoned, top-down changes of the plankton trophodynamics, not just to oil fumes (Crum, 1993; Goldstein et al., 2011; deGouw et al., 2011). Later in 1999, the frequency of asthma in Aberdeen still remained at 24.0% (Devenny et al., 2004), six years after these oil spills and presumed alterations of the plankton state (Scheffer et al., 2001). Farther downstream

in the North Sea after the *Braer* spills, childhood asthma of Norway had increased, from 3.1% in 1981 to 16.1% by 2002 (Carlsen et al., 2006).

In 1995, the prevalences of childhood asthma had spread over a wider area of southeastern England, from the Isle of Wight to Brighton, with a frequency there of 34.1% (Kaur et al., 1998). Later during 2001-2006, deaths of asthmatic English school children expanded downstream along the edge of the North Sea, from Norfolk to Essex Counties, with maximal mortalities also in June-August (Anagnostou et al., 2012), i.e. again during the usual summer HABs of the upstream English Channel (Maddock et al., 1981; Boalch, 1987; Maddock et al., 1989; Vanhoutte-Brunier et al., 2008; Widdicombe et al., 2010) and the Irish/Celtic Seas (Pybus, 1980; Ediger et al., 2001).

Thus, these observations of marine aerosol imports, associated with HABs within coastal waters (Braarud and Heimdal, 1970; Boalch, 1987) of England and Norway, were consistent with high asthma mortality rates along nearshore Counties of Great Britain during 1979-1987 (Higgins and Britton, 1995). Subsequently, by 1999 the prevalence of asthma among school children on the Isle of Wight was 40.3% (Kurukulaaratchy et al., 2003).

The mean rate of asthma prevalence throughout all of the United Kingdom [UK] during 2013 was 9%, with ~1000 UK-wide deaths attributed to this respiratory disease that year. In 1974-1978, the prior UK-wide annual deaths from asthma had been a smaller mean of only ~260 residents yr^{-1} (Charlton et al., 1973). The estimated population of the UK was ~56 million people in 1976, compared to ~64 million residents in 2013, such that the 4-fold UK-wide mortality increment of this pulmonary disease was roughly normalized to the total population base. Given the observed dispersion of the presumed asthma impacts of these marine HAB carriers, what other ill-health consequences might be attributed to them as wind-borne confirmatory evidence?

Recall that the frequency of asthma among school children on the Isle of Wight in the English Channel had increased at an annual rate of 1.1% per year, from 1964 to 1999 (Fig. 5b), attributed to HABs.

4.4 Other medical implications in the North Sea environs

Similarly, in Dublin, Ireland and in Cardiff, Wales, the respective asthma frequencies of those residents were 15.2% in 1995 and 21.6% in 2003 along the western side of the Celtic Sea (Manning et al., 2007), compared to 5.5% in 1973 and 27.3% in 2003 on the eastern side (Burr et al., 1989; 2006). These were similar annual asthma increments of 0.7-0.8 % per year, where HABs of *Gyrodinium aureolum* = *K. mikimotoi* (Fig. 6b) were again present (Pybus, 1980).

Moeover, near Liverpool within the Mersey estuary, the mercury content of surficial sediments had also increased by 9.3% during 1976, compared to 1975, despite reductions of Hg loadings from a nearby chlor-alkali plant during 1974 (Harland et al., 2000), suggesting another source of mercury poisons, i.e nearshore waters of the Celtic Sea. Farther along the English coastline, subsequent DDE/DDT ratios of Welsh breast milk in 1990 ranged from 3.5 to 11.9 (Duarte-Davidson et al., 1994), implying either recent applications of insecticides in some areas, or wind-borne imports of additional legacy marine pesticides.

Previously, large hair mercury amounts (Table 2) of 5.0 ug THg g^{-1} had also been found among residents of Liverpool during 1978 (Sherlock et al., 1982). Furthermore, co-travelling particle sizes of ~5-10 microns of both *Karenia* HABs (Cheng et al., 2005a, b) and sea salt aerosols carried radionuclide tracers of Sellafield origins ~60 km inland from the Cumbrian coastline, at 10 m above ground level over a few hours of sampling during 1979-1980 (Eakins and Lally, 1984; Nelis et al., 1994).

These radioactive, air-borne HAB aerosols may have also explained observations of thyroid cancer increments within downsteam Dublin, since Ireland has no known nuclear facilities, like Australia. Yet, frequencies of thyroid carcinomas within Ireland increased from 19.5% during the 1970s to 37.3% in the 1980s, and then to 43.2% by the 1990s (Dijkstra et al., 2007), as a presumed consequence of the co-transported ^{129}I carcinogens and HAB carriers from near the upstream British and French nuclear waste reprocessing plants at Sellafield and La Hague.

The cumulative ^{129}I loadings of these nuclear waste recycling plants resulted in two orders of magnitude greater amounts of the cancer triggers at those shorelines, than either at the southern entrance of the English Channel, or at the Atlantic boundary of the North Sea (Hou et al., 2002; Michel et al., 2012). Indeed, when HAB carriers were usually present within the English Channel, as during *K. brevis* = *K. mikimotoi* blooms of August 1999 (Llewellyn et al., 2005; Barnes et al., 2015), these vectors also imported radioactive poisons to both French human residents (Pobel and Viel, 1997; Le Guen et al., 1998) and bovines (Frechou et al., 2002; Frechou and Calmet, 2003), like those found in other large mammals (Steinhauser et al., 2012). Indeed, farther downstream in Denmark, the rate of human thyroid cancers there had also increased ~4-fold from 1943 to 2008 (Blomberg et al., 2012; Londero et al., 2013).

Along the French Atlantic coastline, the ^{129}I thyroid activities of cows grazing around a Normandy coastal village, at 3 km away from the La Hague nuclear fuel reprocessing plant, were a maximum of 2080 Becquerels kg^{-1} d.w. thyroid tissue during March 1999 (Frechou et al., 2002; Frechou and Calmet, 2003). By contrast in 1980, when European HABs of *G. aureolum* = *K. mikimotoi* were evidently just restricted that year to the Firth of Clyde (Jones et al., 1982) along the western Scottish coastline of the Irish Sea, the ^{129}I thyroid activities of French cows around

the same sea-spray impacted Normandy village were a ten-fold smaller maximum of 234 Bq. kg^{-1} d.w. during June 1980 (Frechou and Calmet, 2003). Similar radioactive thyroid glands of sentinel cows were also used to describe downwind consequences of nuclear bomb tests within the western United States and Marilinga, Australia (Marston, 1958).

Moreover, within estuarine depocenters of the Seine River, the abundances of sedimentary MeHg stocks increased seasonally ~five-fold at the bottom, from March to September during 1994-1997 (Mikac et al., 1999), coincident with the development of seasonal HABs. Interannual asthma prevalences among residents of the Guernsey and Jersey Islands had also displayed an increment, from 20.6% in 1995 to 25.9% during 2002 (Anderson et al., 2004). Nevertheless, ~50 km inland from the Atlantic coast in Nantes, the hair of French women during 2005-2006 (Pouzard et al., 2010) contained only 0.8 ug THg g^{-1}, despite consumption of fish and at times prior massive HABs (Vanhoutte-Brunier et al., 2008) of *K. mikimotoi*, e.g. during June-July 2003 (Fig. 6).

However, these hair samples were mainly collected during the summer of 2006 (Pouzard et al., 2010), when HABs of *K. mikimotoi* were negligible in the English Channel (Barnes et al., 2015). Thus, without onshore HAB carriers of the mercury aerosols, accumulations of total mercury in human hair were negligible, despite both ~150 g of fish eaten each week per capita (Pouzard et al., 2010) and ~ten French coastal chlor-alkali plants of varying Hg efficiencies.

4.5 Western Mediterranean Seas

Liberated European commensal diazotrophs and HABs, no longer held down by both grazer and nutrient constraints in waters usually warmer than those of the Irish, Celtic, North, Nowegian, and Baltic Seas, also constituted another analog. Off Spain, marine diazotroph precursors, i.e, *Trichodesmium* spp., were studied for more than two

decades (Margalef, 1945; Margalef and Castellvi, 1967), before anthropogenic accelerated arrivals of Saharan dust and emergences of tychopelagic, very toxic dinoflagellates occurred within Catalan waters (Vila et al., 2001).

4.5.1 HABs

As perhaps a harbinger of returning prior fossil coral reefs in these Seas, like those now of the Red Sea, *Ostreopsis siamensis* was rediscovered off Lebanon in 1979 (Abboud-Abi Saab, 1989), with another one, *O. fattorussoi*, recently described from these coastal waters (Accoroni et al., 2016). The Levantine nearshore areas are now mainly more oligotrophic, due to construction of the Egyptian Aswan dams (Oren, 1969; Sharaf El Din, 1977), although sewage outfalls will yield initial successful diatom compeptitors, until silicon-limitations prevail (Taslakian and Hardy, 1976). Accordingly, HABs of both *Karenia* spp. and *O. ovata* were subsequently observed off Alexandria, near the Rosetta mouth of the Nile River, in 1998 (Mikhail, 2001) and 2005 (Ismael and Halim, 2012), once the zooplankton stocks had also declined ten-fold (Zakaria, 2006).

Aerosolized neurotoxins of these HABs recently drove humans away from Italian, French, Spanish, and Algerian beaches, like along other Indian and New Zealand sea shores, with breathing difficulties and hospitalizations (Rhodes et al., 2000; Vila et al., 2001; Brescianini et al., 2006; Zingone et al., 2006; Tichadou et al., 2010; D'Silva et al., 2012; Illoul et al., 2012). In some cases, asthma episodes evidently preceded and/or were associated with concurrent bouts of both bronchial and lobar pneumonia, with deeper penetration of HABs, bacteria, and dust in the latter pulmonary illnesses (Kirkpatrick et al., 2006; Kanatani et al., 2010; Kirkpatrick et al., 2011; Omonijo et al., 2011; Watanabe et al., 2011; He et al., 2012; Kang et al., 2012; He et al., 2013).

Farther to the east, along the Libyan coastline predominatly oligotrophic regions of downwellings usually occurred (Bakun and Agostini, 2001). Before outbursts of *Ostreopsis* spp. were noted along the Algerian coast in July 2009 (Illoul et al. 2012), other *Karenia* spp. HABs (Drira et al., 2008) had prevailed off adjacent Tunisia, since the Libyo-Egyptian WBCs flowed eastward along north Africa, across the mouths of the Nile River, and northward towards Israel, Lebanon, Syria, Turkey, and Greece (Hamad et al., 2005; Gerin et al., 2009).

These nutrient-impoverished waters had a mean NO_3/PO_4 molar ratio of 1.7 in the Bay of Tunis during 1993-1995 (Dalyyhahia-Kefi et al., 2005). Such P-replete nutrient ratios favored more local *Trichodesmium* = *Oscillatoria* spp. (Mounir et al., 2013) precursors of HABs, benefitting from greater iron loadings of an expanded Saharan Desert. Their initial conditions were found near Barbate, Spain (36°8'N, 5°56'W), on the western side of the Strait of Gibralter, as June-July maxima of *T. thiebautii* in 1961-1962 (Establier and Margalef, 1964).

Like the Portuguese clupeoid fishery, Tunisian catches of the zooplanktivore *Sardina pilchardus* had also first peaked by 1965 (Feidi, 1998), when they ate zooplankton herbivores, setting free HABs and other phytoplankton. HABs of *K. sellaformis* and their fish kills were then consequently noted along the Gulf of Gabes during 2005 (Drira et al., 2008). HABs of *O. siamensis* and *O. ovata* had also been recorded in Tunisian waters by 2008 (Mabrouk et al., 2012; 2014).

4.5.2 Consequences

Furthermore, the frequency of asthma attacks among children in the city of Tunis had risen to 15.4% by 2001 (Ait-Khled et al., 2007), compared to 2.5% there in 1968-1970 (Khaldi and Bouyaha, 2001), as another unfortunate human index of altered trophodynamics of the adjacent marine ecosystem. Among residents living next to the Gulf of Gabes, moreover, the

mercury content of their hair was 8.3 ug THg g^{-1} for fishermen, compared to 2.7 ug THg g^{-1} in non-fishers who consumed less fish (Mezghani-Chaari et al., 2011), but in an unknown amount.

Perhaps up to 33% of the total mercury poisoning of these Tunisians may thus have been from inhalation of wind-borne Hg aerosols, transported onshore with the HAB asthma trigger carriers during another trophic cascade. Indeed, considerations next of hair contents of just vegetarians in both Mursin, Turkey and later in Mumbai, India will suggest that perhaps as much as ~40% of sea-side mercury poisons could at times be due to inhalation, rather than just from fish consumption.

Next to a chlor-alkali plant in Mersin, Turkey residents of that sea-side city, who did not eat fish, still had a hair mercury content of 0.43 ug THg g^{-1} during 1996 (Dogan-Saglamtimur and Kumbar, 2010). Presumably, such a Hg hair content was due to onshore dispersions of HAB *Ostreopsis* spp. asthma triggers, found both in surrounding Lebanese (Abboud-Abi Saab, 1989; Abboud-Abi Saab et al., 2013; Accoroni et al., 2016) and Greek (Aligizaki and Nikolaidis, 2006) nearshore waters. Moreover, consideration of additional fish-eaters in Mersin raised their total mean hair mercury content during 1996 (Table 2) to 1.10 ug THg g^{-1} (Dogan-Saglamtimur and Kumbar, 2010), suggesting that as much as ~40% of the mercury poisoning at this Turkish sea-side might have been due to inhalation of HAB carriers of Hg toxins.

Some of the same sequences of malign pulmonary events, without modern loadings of other neurotoxins, had also been observed by the Hippocrates medical group in Greece ~2500 years ago. Then, asthma [Greek word for panting] and pneumonia occurred, when onshore winds blew from the Ionian and Aegean Seas (Jones, 1923). These coastal waters are still sources today of *Karenia* and *Ostreopsis* HABs (Aligizaki and Nikolaidis, 2006; Ignatiades and Gotsis-Skretas, 2010). By contrast, when the dry offshore Miltemis instead prevailed, i.e. Etesian winds from Greek regions inland of Athens, lung maladies were not noted (Jones, 1923) by prior Greek physicians and meteorologists.

5.0 Indian analog

The same sequence of ill events also occurred on the West Indian Shelf [WIS] in the eastern Arabian Sea, upon arrival of both iron-rich dust from African/Indian deserts and mineral aerosols of dessicated Aral Sea origns, subjected to more freshwater diversion projects. Concurretly, first overfishing occurred in a trophic cascade (Vivekanadan et al., 2005; 2009). Then oil spills of tankers and blowouts of the offshore *Mumbai High North* production platform (Sivadas et al., 2008; Rai et al., 2011) reduced (Gajbhiye et al., 1995; Smith and Madupratap, 2005) copepod biomasses (Table 1), while no longer nutrient-limited *Trichodesmium* stimulated growth of ungrazed HABs of *K. mikimotoi* (Iyer et al., 2008; Mahdu et al., 2011). At the mouth of the Indus River, near Karachi, Pakistan, the molar nitrate/phosphate ratios of freshwaters entering the NE WIS were low values of 4-5, at stocks of as much as ~2.0 µmol PO_4 l^{-1} (Harrison et al., 1997), after draining fossil phosphorite deposits, similar to those of Florida.

After relaxation of these regional bottom-up and top-down controls of phytoplankton population dynamics, Indian HABs had increased ten-fold, since 1908 (Padmakumar et al., 2012). Among these blooms along the coastline of SE Kerala State, an accumulation of 57 ug chl

l^{-1} of *K. mikimotoi*, with a diagnostic dissolved N/P ratio of 5.5 to fuel precursor and continuing *Trichodesmium* populations, was observed during 2009 (Madhu et al., 2011).

Prior asthma hospitalizations had also prevailed, both along the Kerala coast during 2004 (Robin et al., 2013) and earlier within the Indian interior at Bangalaru (12 58'N, 77 34'E), along the Western Ghats (Paramesh, 2002). After landward monsoon-driven transports of sea salts (Kulkarni et al., 1982; Adhikary et al., 2007) and HAB asthma triggers ~300 km inland from the coastline of the Arabian Sea, frequencies of seasonal asthma among school children of Bengaluru [Bangalore] had increased from 9% in 1979 to 30% in 1999 (Paramesh, 2002). Over this time period, deaths due to asthma were the largest annual human mortalities, at a mean of 8% between 1966 and 1994 (Ramanakumar and Apajita, 2005).

An aerosol signature of these toxic imports was found in decadal satellite surveys (Dey and Girolamo, 2011; Sreekanth, 2013; Mamun et al., 2014) of the Aerosol Optical Depth [AOD]. Landward dispersion of sea salts and marine HAB asthma triggers, had increased from Arabian Sea sources over 2000 to 2010 during monsoon periods (Dey and Girolamo, 2011), once the disruptive blowouts (Gajbhiye et al., 1995) of the *Mumbai High North* platforms in both 1993 and 2005 had exacerbated an ongoing trophic cascade, due to overfishing (Vivekanadan et al., 2005).

Another radiochemical signature was the 5.3% incidence of thyroid cancer (Fig. 7) in coastal Kollam (8 48'N). Together with the 13.4% prevalence of childhood asthma in nearby Kottayam (9 35'N) during 1994-1995 (Shah et al., 2000), they may have reflected joint aerosol transports from the eastern Arabian Sea of co-mingled sea salts, HABs, and radio-iodine.The long-lived radioactive iodine-129 tracers were released (Snyder et al., 2010) in seawater coolants of three nuclear waste recycling plants (Fig. 7) to co-travel with HAB poisons (brevetoxins +

gymnocins + palytoxins), as they drifted together during seasonal reversals of the West Indian Coastal Current (Shetye and Shenoi, 1988; Shankar and Shetye, 1997; Rao et al., 2008), for subsequent sea spray evasions.

The highest seasonal AOD above Bangaluru occurred during summer (Sreekanth, 2013), when regional computed backward trajectories of air parcels indicated a marine origin (Mamun et al., 2014), particularly during the SW monsoons. Thus, the asthma prevalence was a high amount of 13.4% during 1994-1995 (Table 2) among Indian sea-side residents of Kerala State along the WIS shoreline, but not in Odisha State, next to the diatom-dominated Bay of Bengal. Just an asthma prevalence of 4.0% was found off SE India (Shah et al., 2000) that year (Table 2), i.e. suggesting an asthma contribution of ~9.4% from SW Indian HABs of *K. mikimotoi*.

Accordingly, in the absence of HAB vectors of both asthma and hydragyria, their public health consequences were also minimal in the western Bay of Bengal, where diatoms instead dominated the phytoplankton (Madhav and Kondalarao, 2004; Thangaradjou et al., 2012). The mercury hair content in Bangladesh along the Bay of Bengal was just 0.4 ug THg g^{-1} (Holsbeek et al., 1996), compared to a ten-fold larger arithmetic mean of 5.5 ug THg g^{-1} in Karachi, Pakistan (Ali et al., 2014), along the WIS, where *Trichodesmium* and *Karenia* spp. HABs were the dominant micro-algae. Since the mean hair content of residents in Mumbai [Bombay], India of Maharashtra State was 1.5 ug THg g^{-1} (Yamaguchi et al. 1975), an average value of 3.5 ug THg g^{-1} was assumed for human endpoints of Hg contamination among citizens of Pakistan and western India (Table 2).

By 1995, India was emitting 8% of the global atmospheric loadings of inorganic mercury, from Hg impurities released duing fuel combustion by coal-fired power plants (Pacyna and Pacyna, 2002), with eventual depositions during rainfall to aquatic and terrestrial epicenters of

methylation. About 75% of India's chemical industries were also located in the NW States of Gujerat and Maharashtra, adjacent to the WIS of the eastern Arabian Sea. These commercial facilities included both pesticide factories and brine-requiring original mercury cell chlor-alkali plants.

Before conversions began of about 15 mercury electrolysis plants along the WIS to membrane cell versions during 2003, these prior plant effluents would have earlier added more Hg influxes within Indian rainfalls. Consequently, within Mumbai, the mean hair contents of respectively vegetarians and fish eaters had been 0.8 and 2.1 ug THg g^{-1} (Yamaguchi et al. 1975), so that again ~40% of their mercury poisoning was presumably due to inhalation.

In terms of additional sediment reservoirs, besides storage pools of human scalps, farther downstream in Kerala State, the surface wetlands had an arithmetic mean mercury content of 642 ng THg g^{-1} (Ramasamy et al., 2012), adjacent to the eastern Arabian Sea during 2008. This amount was similar to the mercury contents of some of the SE U.S. soils (Fig. 2) as well, adjacent to the GOM. Moreover, 30% of the total mercury within these Indian near shore sediments was organo-chelated compound (Ramasamy et al., 2012) precursors of MeHg (Bloom et al., 2003; Acquavita et al., 2012). Furthermore, these wetlands had post-monsoon THg and MeHg concentrations that were repectively 76% and 17% of monsoon values (Mohan et al., 2014), implying both seasonal demethylation and onshore aerial imports of these toxic metals to adjacent humans.

Radioactive iodine tracers had been concurrently released from the Trombay and Tarapur nuclear waste reprocessing plants (Snyder et al., 2010), opened during 1965 and 1982 in Maharashtra State, while another recycling facility began operations in Chennai [Madras] of the State of Tamil Nadu, next to the Bay of Bengal, by 1998 (Fig. 7). Thus, the 5.3% incidence of

thyroid cancer (Fig. 7) in Kollam [8 48'N] during 2001-2002 (Nandakumar et al., 2005) and the 13.4% prevalence of childhood asthma in nearby Kottayam [9 35'N] during 1994-1995 (Shah et al., 2000), along the WIS, may have both reflected joint aerosol transports from the eastern Arabian Sea.

Co-mingled sea salt tracers, e.g. long-lived radioactive ^{129}I isotopes from the two WIS nuclear waste recycling plants (Fig. 7), and POPs: Hg effluents released from the many coal-burning power plants and chlor-alkali factories there and modified by nearshore sulphur bacteria; leached pesticides (Kannan and Sen Gupta, 1987); more recent phthalates; and ungrazed (Table 1) HAB DOC carrier poisons (brevetoxins + gymnocins + palytoxins), set free during trophic cascade and oil spill decimations of zooplankton herbivores (Table 1), would have formed a now typical anthropogenic dangerous brew of coastal marine neurotoxins and radioactive mutagens in the eastern Arabian Sea, like most world-wide shelves, adjacent to WCs (Table 2). They would have all drifted south on the WIS during summer monsoons to cause more public health problems of pulmonary disorders and thyroid cancers (Fig. 7) in western Pakistan and India, than along the shorelines of the diatomaceous Bay of Bengal (Table 2).

However, in terms of other neurotoxic phthalate endocrine disruptors, the drainage basin of the Indian Kaveri River begins in the western Ghats of this relatively large CBDP of 3,287,262 km^2. Thus, effluxes of this river into the Bay of Bengal carried HAB-scavenged marine sources of these plasticizers, stored mainly within the WIS. In the Kaveri River, the water concentrations of hormone disruptors were similar to those of the Yangtze River, while the sediment amounts were 7-fold less the Yellow River (Selvaraj et al., 2015). Recall that the Brazilian CBDP was of ~8, 514, 200 km^2 extent, compared to that of 9,596, 920 km^2 for China,

which is the penultimate case of this review. But, next we consider how typical are the results from WBCs thus far.

6.0 South Africa analog

Africa may be the only continent, on which thyroid cancers have not increased much over the last 30 years (Kilfoy et al., 2009; Pellegriti et al., 2013) to provide radiotracers of concurrent POP transfers from the seas to humans. However, *known unknowns* (Rumsfeld, 2011) remain about the environmental and health implications of ~40 year-operations of the SAFARI-1 reactor at the Pelindaba nuclear research facility [25°48'S, 27°56'S] near Pretoria, South Africa, despite fires in 1996 and radionuclide gas leakages in 2009, i.e. like some of the Sellafield accidents. This radio-iodine source is located within the drainage basin of the Crocodile River tributary of the Limpopo River, which in turn reaches the coast of the Indian Ocean, along the shoreline of the western boundary Agulhas Current, south of Mozambique.

Nevertheless, some familiar ominous events had transpired in these coastal waters, which must have involved *labelling* by the same radio-iodine iotopes, if these data were available. For example, neither *Karenia* spp., nor *Ostreopsis* spp. had been observed during nine cruises from 1898-1899 to 1959-1965 within the Mozambique Channel, between Madagascar and Mozambique on the African mainland (Sournia, 1970). Subsequently, the latter tycho-pelagic dinoflagellates of the Indian Ocean, with verified palytoxin poisons (Lenoir et al., 2004), emerged during more recent cruises there by 2007 (Rhodes, 2011; Sa et al., 2013), with already a large asthma frequency of 21.8% observed in nearby Maputo [Lourenco Marques] during 2004 (Mavale-Manuel et al., 2007).

Within the *Trichodesmium*-dominated (Longhurst, 2001; Westberry and Siegel, 2006; Uz, 2007; Poulton et al., 2009; Srokosz and Quartly, 2013) oligotrophy of the more persistent

(Biastoch et al., 1999) Agulhas Current off Durban, South Africa, *Karenia cristata* caused breathing difficulties (Horstman et al., 1991; Botes et al., 2003). The prevalence of asthma was an enormous 50.0% (Jack, 2006) within Durban during 2005 (Table 2). By contrast, downwind of the diatom-dominated EBC of the Benguela Current off Cape Town, South Africa, the asthma prevalence was a smaller amount of 20.3% (Zar et al., 2007), such that 29.7% of the asthma cases may have been caused by those African HABs (Table 2).

Greater pulmonary problems of adjacent humans were known for many years in Durban, compared to residents of Cape Town at the SW coast (Table 2). But, once grazing pressures had been relaxed (Table 1) on the Agulhas Bank of that WBC after 1987 (Verheye et al., 1994; Richardson et al., 2003; Coetzee et al., 2008), subsequent increments of the *K. crisitata* HABs led to greater frequencies of asthma episodes: from <1% of Durban residents during 1963-1967 (Wesley et al., 1969); to 37% by 1998 (Nriagu et al., 1999); and 50% in 2005 (Jack, 2006).

Durban was also located ~45 km downstream of its freshwater drinking supplies, delivered by the Mngceweni River adjacent to a mercury recycling plant at Cato Ridge. There, effluents still led to very large Hg hair contents (Papu-Zamxaka et al., 2010) of 8.3 ug THg g^{-1} by 2007 (Table 2). They were similar to mercury accumulations of 7.0 ug THg g^{-1} (Kinabo, 2005) in hair of residents of coastal Dar Es Salaam, where at least one caustic soda factory was located, near the Agulhas Current. In contrast at Cape Town, next to the diatomaceous Beguela Current, the mercury hair content (Table 2) was just 1.7 ug THg g^{-1} (Airey, 1983).

7.0 Asian analog

Downwind of other expanded Asian deserts, after poor animal husbandry around edges of the Mongolian Desert (Hilker et al., 2014), deposition of iron-rich mineral dust led to 3-fold larger concentrations of dissolved labile iron within surface waters of the Sea of Japan, than in

the western Bering Sea, during 2002-2003 (Takata et al., 2005). Farther to the southeast, in response to earlier upstream aerial Asian dust supplies to the Hawaiian Archipelago (Parrington et al., 1983; Uematsu et al., 1985), nitrogen-fixation within surface waters above the continental slopes had evidently increased ~22% from 1850 to 2000. This conclusion was based upon nitrogen isotope tracers as a second long-term time series stored in long-lived gorgonian gold corals *Kulamanamana haumeaae* (Sherwood et al., 2014). The toxic HAB vector, *O. ovata*, was also dominant among epiphytic dinoflagellates of Hawaii (Parsons and Preskitt; 2007; Parsons et al., 2012).

7.1 The plankton stage

Furthermore, within the Sea of Japan, *Trichodesmium* spp. (Chen, 1972) precursors of dinoflagellate HAB asthma triggers were also present at low N/P ratios of <4 (Kim et al., 2010), compared to most of the East China Sea (Li et al., 2009). Such a phosphorus-replete nutrient ratio would have favored multiple diazotrophic phytoplankton communities (Hashimoto et al., 2012), within contiguous waters of the Tsushima Warm Current in the Korea Strait (Guo et al., 2006), at southern end of the Sea of Japan.

This western boundary current was a secondary branch of the upstream primary Korushio Current, carrying populations of diazotrophic *Trichodesmium* spp. from the South and East China Seas (Minagawa and Wada, 1986; Kao et al., 2001) farther north to the Sea of Japan, as well as to the Yellow, Korean, and Bohai Seas. Diazotroph populations were first reported from the East China Sea during 1972 (Chen, 1982; Zhou et al., 2008), presumably benefiting from recycled phosphorus.

After decomposition of winter diatom blooms, at the seaward edge of Chang Jiang [Yantze] River plume, the spring molar DIN/PO_4 ratios were <10 in May 1998 (Wang et al.,

2003). Moreover, the dissolved mean molar N/P ratio of the adjacent surface Bohai Sea was only 1.1 during 1982-1983, similar to 3.5 in 1960 (Zou et al., 2001; Wang et al., 2003), until ten-fold increased uses of fertilizer nitrogen occurred throughout China between 1970 and 2008 (Zhu and Chen, 2002; Wang et al., 2011).

Finally, *Ostreopsis* spp. had penetrated Russian waters past the Sea of the Japan by 2009 (Selina and Orlova, 2010; Efimova et al., 2014), while *Karenia* spp. HABs off China had initiated diagnostic massive kills of caged fish earlier in 1988 (Young and Hodgkiss, 2004). Before then, HABs of *K. mikimotoi* had also grown within Japanese waters, from small amounts during 1934-1935 to frequent "*red tides*" begun in 1961 (Okaichi, 2004), while carrying ashore methyl mercury poisons to be recorded in human wigs (Suzuki et al., 1984).

7.2. The same plot

Like other regions discussed here, these Asian HABs served as carriers of additional organic neurotoxins, besides the reported episodes of both asthma and pneumonia (Kanatani et al., 2010; Kang et al., 2012). Ten-fold decadal increments of asthma attacks had occurred in Tokyo, from prevalences of 0.7% there in 1965 to 14.0% in 1996 (Baba et al., 1966; Futamura et al., 2011). Similarly in Shanghai, asthma occurrences increased from 1.8% in 1990 to 10.2% by 2012 (Asher et al., 1998; Huang et al., 2015).

HAB onsets also began in Asian coastal waters the same way over the last half-century, due to both greater nitrogen-fixations and fewer herbivores. The changed top-down forcings were in usual responses to regional overfishing, oil spills, and dispersal of nuclear wastes from local recycling plants in China and thoughout Japan. Again these latter activities also provided radiotracers of onshore co-transports of halides (Furukawa et al., 2013) and HAB-carried neurotoxic poisons.

Within the Sea of Japan, between Korea and Honshsu, the annual abundances of copepods had declined ~10-fold after 1990 (Iguchi, 2004). Little recovery of zooplankton stocks then ensued from 1991 to 1999 (Kang et al., 2002; Lee et al., 2009), once local trophic cascades maintained the initial increment of top-down predation demands by the zooplanktivore Japanese sardine *Sardinops melanostictus* since the 1980s (Ohshimo et al., 2009). Similar 40% declines of zooplankton had been found in the Bohai Sea between 1959 and 1992 (Tang et al., 2003). Finally, decimations of herbivores were noted (Table 1) within the intervening Yellow Sea during 1959-1993 (Lin et al., 2001).

Century-long accumulations of methyl mercury [MeHg] within the presumed air-borne organic poison carriers of *Karenia* brevetoxins and *Ostreopsis* palytoxins were also stored in Japansese female wigs. Indeed, ten-fold increments in the Hg contents of their hair, upon assumed inhalation, were estimated between 1880 and 1970 (Suzuki et al., 1984), with an arithmetic total mercury mean of 6.0 ug THg g^{-1} found in 1965 (Nakagawa, 1995).

On the predominantly lee side of the Sea of Japan, after **1**) marine Hg entrainment by aerosolized HAB carriers, **2**) wet deposition (Sakata and Marumoto, 2005), and **3**) inhalation, the arithmetic mean of human hair on Shikoku Island, Japan was 2.5 ug THg g^{-1} during 1975-1976 (Shimomura et al., 1980). While in Korea, along the western side of the Sea of Japan human hair during 2005-2006 contained only 0.9 ug THg g^{-1} (Kim et al., 2008).

In our estimate of the contemporary Japanese mercury poisonings (Table 2), a residual hair content after 9 years of still 23.6 ug THg g^{-1} among the Minimata Bay victims (Yamaguchi et al. 1975) has been ignored by us, lest the consequences of wind-borne HAB poisons be overstated. Gaseous mercury is still evaded from that Bay (Marumoto and Imai, 2015), for example. Yet, once red tides of *Karenia* spp. had peaked in downstream Hong Kong, the human

hair total mercury contents there stabilized at 3.1 ug THg g^{-1} in 1982 (Airey, 1983) and 3.6 ug THg g^{-1} in 1995 (Dickman et al., 1998), i.e. after the massive fish kills in 1988. Recall that the WHO proposed limit for total mercury [THg] is 2.5 ug THg g^{-1} [ppm] in human hair samples (Bellanger et al., 2013).

7.3. The unfortunate radio-tracer

In terms of radio-iodine tracers, arrivals of mineral aerosols in the environs of the Bohai and Yellow Seas, as well as in the adjacent Sea of Japan, were first reported during 1150 B.C. as "*dust rain*" by Chinese observers (Sheng et al., 1981), with far-field origins, which included the Loess Plateau silt deposits of northwest China. From between Lanzhou [Lanchow], along the banks of the Huang He [Yellow] River, and Beijing [Peking], at the head of the Bohai Sea, such dusts moved at different altitudes of varying atmospheric circulations over the last ~30,000 years of the Pleistocene and Holocene periods (Pye and Zhou, 1989).

Along the bank of the upstream Huang He River in Lanzhou, a uranium conversion plant opened in 1980, with subsequent release of radio-iodines into the downstrean Bohai Sea. Exported sea sprays and HAB-lysed DOC carriers during the SW monsoons would have carried thyroid cancer triggers to interior NE regions of China, such as Heilongiang Province. There, the capital city of Harbin had a two-fold larger frequency of thyroid cancers during 2008-2009, than in the United States (Hegedus, 2004; Yang et al., 2011). To complete the onshore cycle of this CPPD, rainfall served as a phthalate vector to adjacent Chinese residents, upon wet deposition to freshwater reservoirs of Harbin (Liu, et al. 2013).

With seasonal wind reversals to the NW monsoons, the transformed aerial radio-iodine effluents from Lanzhou, China would have been joined with more biotic-harvested sea sprays of I^{129} from at least two other contemporary Japanese nuclear waste reprocessing plants. The

Tokaimura plant was opened on the east coast of Japan, north of Tokyo during 1977, whereas the Rokkasho plant was commissioned in 2005 at the northern end of Honshu Island, along Tsugaru Strait between Hokkaido and entrance to the Sea of Japan. Farther south along the shorelines of the East China Sea, onshore wind-borne aerosol co-transports of HAB toxins and radiohalides would have contributed to the observed 2-3 fold increments of thyroid cancers among adjacent humans of Chinese coastal population centers.

The ionizing radiation sources of iodine isotopes were presunably ferried ashore there by the marine DOCs (Duce et al., 1963) of HABs, like those described thus far in Brazil, South Carolina, Ireland, France, Denmark, India, Japan, and soon among Australian residents. Thyroid carcinomas increased from a mean fequency of ~1% of all neoplasms in 1983 to ~3% in 2007 at Shanghai (Wang and Wang, 2012). Similarly, in Hong Kong, the mean prevalence of thryroid cancers also grew from ~3% of both men and women during 1983 to ~6% in 2007, across all age groups (Xie et al., 2014), suggesting a common environmental factor, rather than human aging.

7.4. The penultimate denoument

Applications of endocrine-disrupting pesticides in northwest Asia had declined since 1972 as well, based upon DDE and DDT contents of human breast milks. Within Osaka, Japan this ratio of female residents was 3.1 in 1972, compared to 27.7 in 1994 (Konishi et al., 2001). Similarly in Beijing, China, the DDE/DDT ratio of lactating women increased from 7.2 in 1982 (Yu et al., 2005) to 19.8 in 2009 (Wang et al., 2011). Yet, like in Brazil, these neurotoxins were replaced by those of phthalates in China, Japan, and other areas of the western Pacific Ocean. Along the east coast of Australia in the State of Queensland, the *p,p'* DDE metabolite of DDT pesticide residues was also 12800 ng DDE g^{-1} fat within Brisbane residents in 1973 (Miller and

Fox, 1973), compared to 255 ng DDE g^{-1} fat there in a ratio of 25.5 during 2003 (Mueller et al., 2008).

The new endocrine disruptors of high-molecular-weight phthalate esters DEHP and their major metabolite mono (2-ethylhexyl phthalate [MEHP], found in human urine, were associated with household dust in France (Bonvallot et al., 2010) and Japan (Bamai et al., 2015). Inhalation of DEHP also amounted to as much as 47% of the intake of phthalates by workers within industrial environments in Taiwan (Fong et al., 2014).

Thus, the same Asian desert mineral aerosols that carried iron supplies to *Trichodesmium* would have co-travelled with: **1**) ionizing radiation triggers of papillary thyroid cancers (Richardson, 2009; Meredith et al., 2014), of high cure rates; **2**) HAB asthma vectors; **3**) air-borne sources of hydragyria; and **4**) alternating DDT and phthalate endocrine perturbants. Accordinly, the urine content of MEHP in residents of Hokkaido, Japan was a larger 20.7 ug l^{-1} in 2010 (Bamai et al., 2015), compared to 6.0 ug l^{-1} among Australians in Queensland during 2013 (Gomez Ramos et al., 2016), with a local dust supply from the Simpson Desert, and just 3.3 ug l^{-1} released by inhabitants of Puerto Rico during 2012 (Cantonwine et al., 2014).

8.0 Oceania analog

In air above the last case of the oligotrophic East Australian Current [EAC] within the Coral and Tasman Seas, westerly Australian dust storms (McTainsh et al., 2005; Leslie and Speer, 2006; Mackie et al., 2008) exited the east coast. Due to continued drought conditions, the dust storm in Brisbane during September 2009 was the worst of the previous 70 years. Yet, these dust loadings continued to fuel Fe-starved *Trichodesmium* populations on the adjacent Great Barrier Reef. After local increments of these diazotrophs since 1928 (Bell et al., 1999),

Ostreopsis HABs had increased two-fold on the Great Barrier Reef between 1985 and 2007 (Skinner et al., 2013).

8.1 Ill health impacts

The coral cover of this Reef also declined by ~50% between 1985 and 2012 (De'ath et al., 2012), concomitant with increases of both *Ostreopsis* and *Gambierdiscus* spp. A resident of Brisbane died from ciguatera, after eating sawtooth barracuda *Sphyraena punamiae* in 2008 (Hamilton et al., 2010), of presumed *Gambierdiscus lapillus* origins (Kretschmar et al., 2016). Before this recent event, the last recorded case of Australian ciguatera was again in Brisbane during 1967 (Clark and Whitwell, 1968). Like Gulf Stream poleward exports of barracuda and ciguatera into the SAB, southward movements of the EAC also led to apperances of *Gambierdisucs carpenteri* off southern New South Wales in 2013 (Kohli et al., 2014).

Prevalence of asthma among residents of Brisbane had also exhibited a 4-fold increment over a longer time period, from 6.1% during 1959-1968 (Derrick, 1972) to 28.4% by 1982 (Burgess et al., 2006). In Sydney, the frequency of childhood asthma concurrently increased ten-fold, from 2.7% in 1952 to 28.5-30.0% during 1991-1993 (Solomon, 1952; Pearce et al., 1993; Robertson et al., 1998). Such a toxic HAB stimulus of malign human pulmonary events followed both overfishing-induced trophic cascades and anthropogenic coral bleachings (Hughes, 1994; Hughes et al., 2003), as well as the increments of local dust-fed, bottom-up diazotroph nutrient sources from the interior deserts.

Before replacement of prior pesticide endocrine disruptors by Australian phthalates, the ratios of DDE/DDT in human breast milk were means of 12.8 in the State of Victoria and 32.4 over the rest of Australia, i.e. suggesting more frequent applications of insecticides along the SE coastline. Like Amazonian Brazil, severe hair mercury contaminations of 21.9 ug THg g^{-1}

occurred in gold mining areas (Abe et al., 1995) of Lake Murray, New Guinea. Whereas, the mining legacies of gold extractions in Australia were smaller hair mercury amounts of 2.7 ug THg g^{-1} in Darwin and 2.2 ug THg g^{-1} in Cronulla (Airey, 1983), i.e. approximating those of the WHO guideline.

8.2 Enigmatic radiotracers

Finally, in terms of radio-tracers of concomitant pollutant transfers, the iodine sources of ionizing radiation, reflected in prevalences of thyroid cancers, are still the subject of heated debates in a presumably nuclear-free economy of Australia. Yet, the incidences of thyroid cancer among Australian adults of New South Wales, increased from 1.6% during 1973-1977 to 5.3% in 1998-2002 (Kilfoy et al., 2009), despite the absence of Australian civilian nuclear waste reprocessing and power plants (Henningham, 1996; Pfingsten et al., 2001; Danesi et al., 2002; Yamada, 2004; Willis, 2006), while holding ~33% of the world's uranium deposits.

Like *G. lapillus, G. carpenteria,* and *G. yasumotoi* (Rhodes et al., 2013), the other extant, but aerosolized, dinoflagellate HABs of *Ostreopsis siamensis* (Rhodes et al., 2000; Chang et al., 2000) and *Karenia papilionacea*, *K. selliformis, K. brevisulcata*, and *K. bidigitata* (Haywood et al., 2004) would have drifted polewards within the EAC. Thus, thyroid cancer mutagens could also been carried ashore by these latter HABs towards both SE Australia and New Zealand, across the Tasman Sea, but from what radio-iodine sources?

There had also been relatively small releases of local Australian medical, short-lived iodine radio-wastes in sewage outfalls of Sydney (Carolan et al., 2011). But, given the long half-life of ^{129}I, two more likely options are: prior British nuclear bomb tests at the Montebello Islands, Emu Field, and Marilinga in Australia; and French nuclear bomb tests at Moruroa Atoll in French Polynesia. During 1952-1957, 12 bombs were exploded along the west coast, and

within the interior, of Australia, with wind-blown dispersions of local fallout monitored by subsequent radioactive thyroids of cattle (Marston, 1958). The French followed, with ~198 bomb tests over 1966-1996 on Moruroa, at 1250 km upstream of Tahiti.

8.0 *In situ* marine teleconnections

Downstream at Tahiti within the South Equatorial Current [SEC], moreover, 11.5-14.3% of the schoolchildren were asthmatic during 1979-2000 (Perdrizet et al., 1987; Liard et al., 1988; Foliaki et al., 2007), reflecting possible marine HAB carriers of radioiodine (Duce et al., 1963). After the French bomb tests on upstream Moruroa and Fangataufa Atolls [21°50'S, 135°50'W] of the SEC during 1966-1975, a Tahitian hospital at 17°40'S, 149°25'W subsequently reported 2-3 fold larger incidences of thyroid cancers by 1985-1995, compared to similar ethnic populations of Hawaii and New Zealand (de Vathaire et al., 2000).

Asthma prevalences in Suva, Fiji, were also 18.0% during 2002 (Foliaki et al., 2008). Around these islands [18°S, 179°E], commensal HABs of at least *Ostreopsis* spp. (Skinner et al., 2011) were fueled by local colonial diazotrophs (Dupouy et al., 2011). Indeed, the type locale of *Gambierdiscus toxicus* was in the Gambier Islands (Adachi and Fukyo, 1979), ~500 km away from the Tuamotu Archipelago, where the French nuclear tests were performed.

Hoever, by 1998, the incidences of ciguatera in both French Polynesia and Fiji had become the same ~2% (Skinner et al., 2011), with the implication that the importance of the original physical destruction of the latter bombed coral reefs had been overriden by **1**) ubiquitous thermal bleachings of dinoflagellate benthic habitats and **2**) diminished (Table 1) copepod herbivores, including the local radio- poisoned ones (Jeffree et al., 1997), no longer eating all of either the pelagic, or tychopelagic, HABs.

For example, the bio-accumulation of ^{137}Cs was 13-fold for *Calanus helgolandicus* (Polikarpov, 1966), in relation to dissolved concentrations of this seawater radionuclide (Bowen et al., 1971). All marine plankton accumulated cesium, as if it were potassium required for biotic metabolic processes involving the latter (Williams and Swanson, 1958). Moreover, the ^{137}Cs contamination around the Moruroa and Fangataufa test sites was ~5-fold greater in 1972, than over areas of French Polynesia (de Vathaire et al., 2000).

Finally, within the Fijian Archipelago and on downstream New Caledonia [21^{o}25'S, 165^{o}30'E], where poisonings of *Gambierdiscus* spp. impacted ~35% of the residents in 2005 (Baumann et al., 2010), the human thyroid cancer rates (Ballivet et al., 1995; De Vathaire and Le Vu, 1996; De Vathaire et al., 2000; Truong et al., 2007), were some of the highest reported by the International Agency for Research on Cancer [IARC]. But, any linkage to earlier French bomb tests on Moruroa Atoll of the SEC was initially dismissed, based on the short 8-day half-life of just ^{131}I and travel times of associated upstream air parcels to Tahiti (De Vathaire et al., 2000).

Yet, following these nuclear explosions in French Polynesia during 1966-1975, the ^{129}I inventory within downstream corals of the Solomon Islands [8^{o}S, 159^{o}E] was eventually measured (Biddulph et al., 2006). As annual indices of changing water column concentrations there, the ^{129}I Solomon accumulations had increased five-fold between 1975 and 1995, tracking influxes of bomb-contaminated seawater. By contrast, the levels of radio-iodine activity of ^{129}I within upstream Easter Island corals remained nearly constant (Biddulph et al., 2006), reflecting just previous global fallout increments of earlier farfield global bomb tests.

Within mean annual westward flows of ~20 km day^{-1} in the SEC at 110^{o}-140^{o}W (McPhaden et al., 1991), and between 10^{o}-30^{o}S (Reid, 1997; Webb, 2000), it would have taken

only a few months for surface waters, containing dissolved radionuclides of ^{129}I and impacted plankton carriers to move downstream 1000-2500 km towards the Solomon Islands, Papua New Guinea, Australia, and New Zealand. Even the relatively short half-life of ~30 years for radiative decay of ^{137}Cs would still have been an effective trophic perturbant en route, relaxing grazing pressures.

Thus, the eventual downstream air-borne marine aerosols of SEC sea sprays, pushed against windward atolls and continents, would have carried ashore radioactive sea salt ^{129}I stimuli of a very long, ~16 million-year half-life. Upon arrivals, these radio-tracers would have presumably both caused thyroid cancers and *labelled* concurrent asthma, mercury poisoning, and ill-fated endocrine events of therlache and cryptorchidism disruptions within Oceania, and elsewhere. Whereas, ciguatoxins evidently just remained behind in sea water, since by contrast aerosolized palytoxins have yet to be also found in parrotfish vectors (Suzuki et al., 2013).

9.0 Conclusions

Based on our review here and a global estimate of 300 million world-wide asthmatics during 2004 made by the Global Initiative for Asthma [GINA] program (Masoli et al., 2004), we suggest that ~45 million persons suffered asthma attacks of marine HAB origins that year. Of those victims, 33,000 people died, assuming again that HABs caused 15% (Table 2) of the world-wide asthma deaths that year (Masoli et al., 2004). In this process, as much as 40% of mercury poisonings may instead have been effected by inhalation of collateral HAB-carried marine neurotoxic aerosols of MeHg, not just from eating marine fish. Additional ill-health maladies of human endocrine disorders would have resulted from accompanying wind-borne onshore fluxes of legacy pesticides and phthalate esters, stored temporarily nearby in shelf sediment depocenters, before resuspensions within storm-induced sea sprays. In some regions,

next to past bomb sites and present nuclear waste recycling and power plants, radio-iodine tracers of varying half-lives would have elicited thyroid carcinomas as well.

Aerosol penetration extents of sea salts are known over distances of >500 km inland from both the GOM (Junge and Gustafson, 1957) and the eastern Arabian Sea (Adhikary et al. 2007), depending upon the strengths of daily sea breezes, episodic cyclones, and seasonal monsoons. Each human within those onshore marine aerosol fields may have breathed ~20,000 liters day^{-1} of air, potentially contaminated by HAB, MeHg, DDT/DDE, and, phthalate POP neurotoxins.

In each of the 10 paired EBCs and WBCs analyzed here, the human hair endpoints of mercury contamination were greater next to WBCs, where HABs prevailed, yielding a five-fold larger arithmetic mean than other sea-side residents, living next to EBCs (Table 2). Thus, the WFS was an analog of nine additional overfished coastal ecosystems, adjacent to oligotrophic WBCs, where trophic cascades, in addition to POP impacts, decimated copepod herbivores (Table 1).

Upon regional desertifications, the WBCs were dominated by the recently Fe-fertilized colonial nitrogen-fixers, *Trichodesmium* spp., in contrast to the diatom-rich EBCs. Century-long nitrogen isotope (Christensen and Richardson, 2008; Sherwood et al., 2014) and hair mercury (Suzuki et al., 1984) time series within world-wide coastal regions have shown that both commensal nitrogen fixers and toxic HAB carriers of algal and mercury organic aerosol toxins have all increased, in response to multiple global anthropogenic perturbations. More long-term times series of iodine isotope ratios in corals (Biddulph et al., 2006) have also documented the unsuspected large spatial scales of pollutant transfers via ocean curents, rather than just known atmospheric teleconnections (Namais, 1972).

Finally, next to these and other coastal ecosystems, world-wide increments of breathing attacks among school children were documented by 342 multi-decadal ISAAC surveys of asthma prevalence, within 56 countries (Fig. 4). Consequently, physicians know that global asthma attacks, as well as papillary thyroid carcinomas, are increasing on a planet covered by ~70% seawater, but they do not know why. Whereas, marine scientists know that deserts have expanded, coastal waters are eutrophied, food webs are overfished, oil spills happen, heavy metals poison fish and air, while HABs are also increasing, but they do not know the extent of those pressing public health impacts, which afffect them and their families.

These are not new problems, since inhalations of mercuric aerosols in South American mines (Lombardi et al., 2012; Avila et al., 2014) were major sources of mercury poisoning, including cinnabar extractions by Zapotecs during the same time period of wind-driven malign pulmonary triggers, studied in ancient Greece (Jones, 1923). Past knowledge of the recognized ill-health responses to inhaled Hg aerosols were the persistent political decisions to use prisoners in the Almaden mercury mines of Spain, both from 1583 to1801 and earlier during Roman times.

Yet, this review suggests that continuing ill health consequences for global human societies of air-borne neurotoxins have expanded, outside of mines and downwind of ongoing industrial facilities, to coastal seas. The aerial returns of combined "legacy" mercury pools, stored temporarily in present coastal seas (Amos et al., 2013), together with those of similar DDT/DDE (Stemmler and Lemel, 2009) and phthalate ester (Peijnenburg and Struijs, 2006) neurotoxins, can no longer be ignored, nor the pulmonary consequences of their HAB carriers (Walsh et al., 2015; 2016; 2017).

Obviously, solutions to these problems of declining global zooplankton abundances (Table 1) and HAB increments are required in relation to ongoing ISAAC and GINA

documentations of increasing world-wide pulmonary diseases and perhaps unrecognized wind-borne vectors of mercury poisonings (Table 2). But, one must first place these ecological and public health crises in the "*appropriate setting*" of a larger holistic context (Fig. 4). Rather than just another "*consilience*" of science and the humanities (Wilson, 1998), or of fishery scientists and oceanographers (Pauly, 2001), it is now time for a total new "*paradigm shift*", requiring a marine eco-medical consilience for our planet.

Indeed, because of the continued insularity (Pontecorvo, 2003) between oceanographers and fishery biologists, let alone among economists, medical doctors, statisticians, and environmental academicians, barriers of disciplinary mind-sets remain. Heated debates about the statistics of cancer and cigarette smoking (Doll and Hill, 1950; Fisher, 1958a, b , c) were harbingers of still inadequate data sets and naïve causal models today. They plagued contemporary and subsequent natural resource managers (Cushing, 1974; Larkin, 1977), as well as present ones (Wagener, 2005; Wardlaw et al., 2006). Medical and environmental science communities should expand working together to provide **1**) eventual remedial actions for poison reductions at the sea sources, and **2**) immediate provisions of public health warnings to adjacent humans of urban and rural areas. Then both prudent avoidances of aerosols and stockpiles of needed drugs, within >500 km of the ocean boundaries of afflicted developing and developed nations, would follow.

Time will tell whether repeatedly impaired growth of the dominant marine copepods had been allowed so few respites between their past population losses (Table 1), such that their resiliances were compromised (Scheffer et al., 2001; Scheffer and Carpenter, 2003). Given multiple causes of these zooplankton decrements (Table 1) and HAB-mediated increments of adverse public health responses (Table 2), future remediation efforts will also require solutions

effected by many sectors of society. Yet, past results of prior models (Walsh et al., 2017), together with the present review of those enormous validation data sets embodied in just ten known coastal WBC analogs, show that sufficient early numerical model warnings are a feasible goal. All sea-side nations would improve their medical care of continuing victims of inhaled marine poisons, once proven hurricane predictions of physical landfalls and aerosol influxes are coupled to other forecasts of biotic consequences, ranging from phytoplankton to people.

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Table 1. Decimation of world-wide zooplankton stocks within continental shelves

REGION	CAUSE	DATE	REFERENCE
Plymouth, England	Cascade, Oil	1925-1994	Southward et al., 1995
West Indian shelf	Cascade/DDT/Oil	1933-1994	Kannan & Sen Gupta, 1987; Gajbhjiye et al., 1995; Smith & Madhupratap, 2005; Vivekanadan et al., 2005; 2009
Enewetak Atoll	Radioactivity	1946-1955	Johnson, 1954; Gilmartin, 1958
West Florida shelf	Cascade	1949-1979	King, 1950; Khromov, 1969; Walsh et al., 2011; 2015
Southwest Brazil shelf	Cascade, Oil	1950-2004	Domingos-Nunes and Resgalla, 2012; Zanardi-Lamardo et al., 2013
Egyptian shelf	Aswan Dam	1962-1966	Zakaria, 2006
California Current	Oil	1951-2006	Roemmich and McGowan, 1995; Mackas and Beaugrand, 2010
Bering Sea	Cascade	1954-1994	Sugimoto and Tadokoro, 1997
Adriatic Sea	Cascade, Hg	1959-2005	Berline et al., 2012
Yellow Sea	Cascade, Oil, Hg	1959-1993	Lin et al., 2001
Gulf of Thailand	Cascade, Hg	1962-1998	Brinton, 1963; Jivaluk, 1999
Peru Upwelling	Cascade	1964-2002	Ayon et al., 2008
Bay of Campeche	Cascade, Oil	1965-1981	Khromov, 1969; Guzman del Proo et al., 1986; Walsh et al., 2015
Villefranche	Cascade, Oil	1966-2003	Berline et al., 2012
Gulf of Guinea	Cascade, Oil	1969-1992	Wiafe et al., 2008
Sea of Japan	Cascade	1973-1995	Iguchi, 2004
Black Sea	Cascade	1976-1990	Daskalov et al., 2007
Gulf of Maine	Cascade, Oil	1977-1990	Kane, 2007
Baltic Sea	Cascade, Oil	1977-1996	Mollmann and Koster, 2002
Barents Sea	Cascade	1984-1992	Dalpadado and Skjoldal 1996
South China Sea	Cascade, Oil	1984-1993	Yin et al., 2006
Naples	Cascade, Oil	1984-2006	Berline et al., 2012
Saronikos Gulf	Cascade	1987-2004	Berline et al., 2012
Normandy, France	Cascade, Oil	1987	Samain et al., 1980
West Agulhas Bank, SA	Cascade	1988-2003	Huggett et al., 2009
Mallorca	Cascade, Oil	1994-2003	Berline et al., 2012

Table 2. Relative contributions to asthma episodes from HABs within coastal regions, adjacent to paired western [WBC] and eutrophied [EBC] boundary currents, with 6-fold less mercury content [ppm] of human hair found (year) in EBCs. The other top data entries of column four are human total asthma frequencies (%) surveyed {year} in the presence of both HABs and other asthma triggers, e.g. dust and chemical pollutants, of WBCs. The middle entries of this column are contrasting prevalences of total asthma conditions in absence of HABs, when diatoms instead dominated EBC phytoplankton communities. The last bottom entries of the fourth column are thus the differences of these asthma-inducing triggers, or those inferred regional contributions of coastal dinoflagellate HAB carriers of their poisons and other organic neurotoxins [MeHg and DDT] and radionuclides [^{129,131}I] during asthma attacks. After subtraction of other stimuli, a global HAB mean of ~15% prevailed during sampling periods of 1978-2013, as the world-wide average of the entries of column 5.

1. Region	2.Year	3. Mercury	4.Total asthma	5.Percent due to HAB
TX-NC, SC SE U.S. - WBC	(2001, 2007), {2003}	1.1, 2.7	9.6	
Southern California – EBC	(1999), {2008}	0.4	6.4	
			3.2	3.2%
Mersey, Isle of Wight, - WBC	(1978), {1999}	5.0	40.3	
Murtosa, Lisbon Portugal - EBC	(2000), {2000}	1.1	15.0	
			25.3	25.3%
Tunis and Sidi Mansour - WBC	{2001}, (2009)	2.4	15.4	
Egypt, Cairo Nile River - EBC	{1991}, (2000)	0.2	8.0	
			7.4	7.4%
Mersin, Turkey - WBC	(1996), {2004}	1.1	17.8	
Sicily and Slovenia - EBC	(1978), {2000}	0.3	11.8	
			6.0	6.0%
Rio de Janeiro, Brazil -WBC	(2004), {2012}	3.6	23.7	
Concepcion, Santiago Chile - EBC	(1994), {1994}	0.4	13.9	
			9.8	9.8%
Kottayam – Karela, India -WBC	(1984), {1995}	4.3	13.4	
Chennai, Bay of Bengal - EBC	{1995}, (2011)	0.4	2.1	
			11.3	11.3%
Kuwait, Arabian Gulf - WBC	(1995), {1995}	4.1	16.8	
Dakar, Kedougou Senegal - EBC	{2012}, (2013)	1.1	3.0	
			13.8	13.8%
Durban, S. Africa -WBC	{2005}, (2007)	8.3	50.0	
Cape Town - EBC	(1982), {2002}	1.7	20.3	
			29.7	29.7%
Tokushima, Japan - WBC	(1996), {1996}	4.6	14.0	
Portland, Oregon - EBC	{1995}, (2007)	0.6	3.0	
			11.0	11.0%
Darwin, Brisbane - WBC	(1982), {1984}	2.5	28.4	
Papua New Guinea - EBC	{1984}, (1988)	0.8	1.3	
			27.1	27.1%

List of Figure Legends

Figure 1. Annual chronic lower respiratory disease mortalities, including asthma + pneumonia deaths = 28% in Florida during 1970-2013 (solid line) and in Georgia during 1994-2013 (dotted line), with each set of total death rates adjusted to the U.S. standard population of 100,000 residents (Data courtesy of the Florida Department of Health and the Georgia Department of Public Health). Other lung fatalities were due to influenza, bronchitis, emphysema, and chronic obstructive pulmonary disease [COPD].

Figure 2. Total mercury (ppb) in surface soils of the southeastern United States during 1961-1970, in relation to the sums of pneumonia and influenza deaths within the same 1027 counties, from Texas to Virginia during 1999-2011, compiled mainly by the U.S.G.S. and C.D.C. Note the apparent onshore tongues within the >150 ppb isopleth of presumed wind-borne marine MeHg components, methylated in estuarine hypoxic surficial sediments.

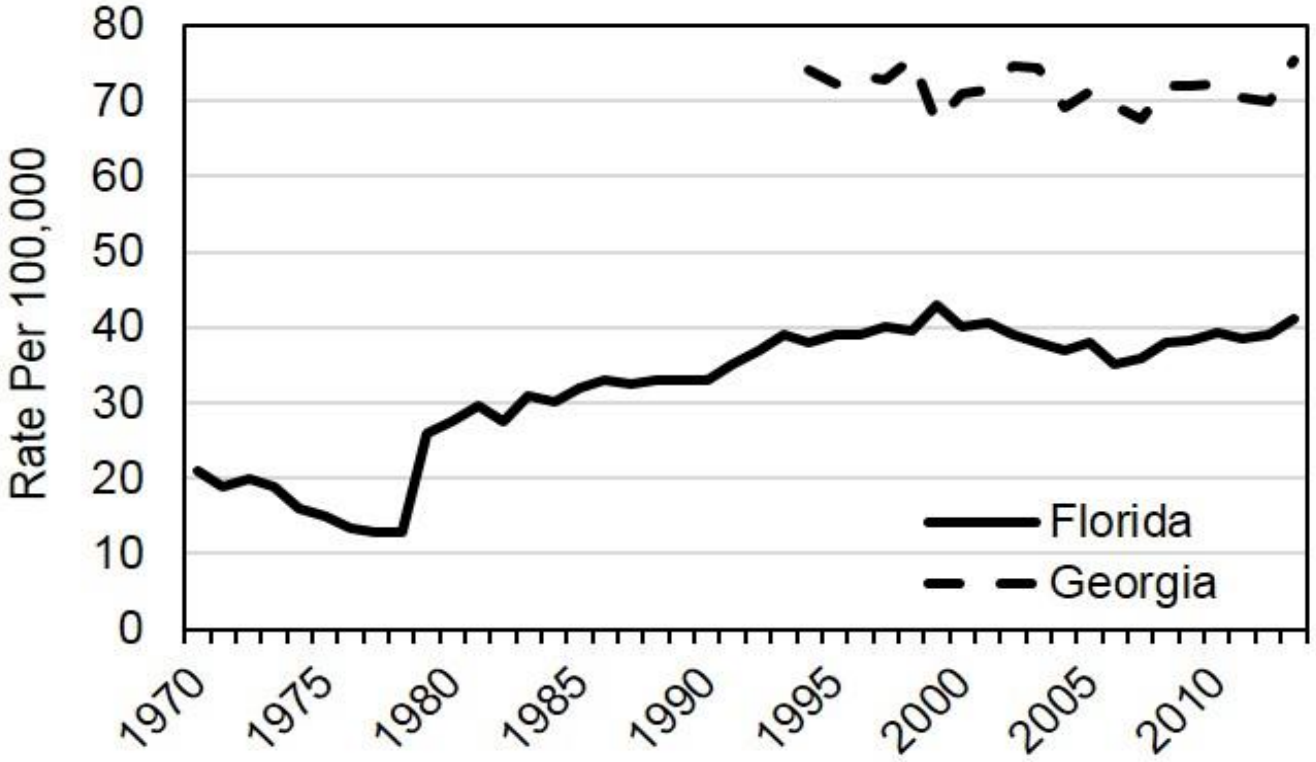
Figure 3. Parallel satellite estimates of chlorophyll concentrations of total phytoplankton within both sides of the Gulf of Mexico, using the same CZCS sensor over time, from composites of (a) October 1979, with respect to seasonal time series on (b) WFS and (c) BOC shelves as indicated by the red arrows, over a trophic cascade of 1978-1986.

Figure 4. Multi-year concurrent studies of 258 ISAAC surveys in 1958-2004 of coastal asthma prevalence of children in England, Scotland, Norway, France, Morocco, Egypt, Italy, Kuwait, Pakistan, India, Malaysia, Thailand, Hong Kong, Japan, Brazil, Barbados, Australia, New Zealand and the United States, downplume of Fe-rich, desert dust (black arrows). Asthma prevalence of >10% and concurrent mercury and DDT poisonings by HABs of *Karenia* spp. vectors are indicated by the red symbols, whereas more potent dinoflagellate *Ostreopsis* vectors are the green symbols. When nuclear power and waste recycling plants were situated in coastal regions, adjacent thyroid cancers also prevailed in those areas.

Figure 5. The vertical lines of the A) malign pulmonary events of tropical/subtropical regions are coral bleachings over the last half-century, whereas those of the concurrent B) European asthma time series are oil spills.

Figure 6. Distributions of MODIS satellite-sensed populations of A) *Trichodesmium* and B) *Karenia mikimotoi* within the English Channel environs during 9-10 July 2003.

Figure 7. Prevalences of thyroid cancer during 2001-02 (after Nandakumar et al., 2005) in relation to the Trombay, Tarapur, and Kalpakkan nuclear waste reprocessing plants opened in 1964, 1982, and 1998. Two recycling facilities were north of Mumbai in Maharashtra State, near the West Indian Shelf, and the third was at Chennai, near the Bay of Bengal (red stars). Presumably, I-129 and I-131 radionuclides must have been released to both coastal regions. Sea salts in air above NW Gujarat and SW Kerala States were similar, but commensal HABs of *K. brevis* and *K. mikimotoi* occurred south of Goa, after upstream N-fixation by *Trichodesmium* precursors.



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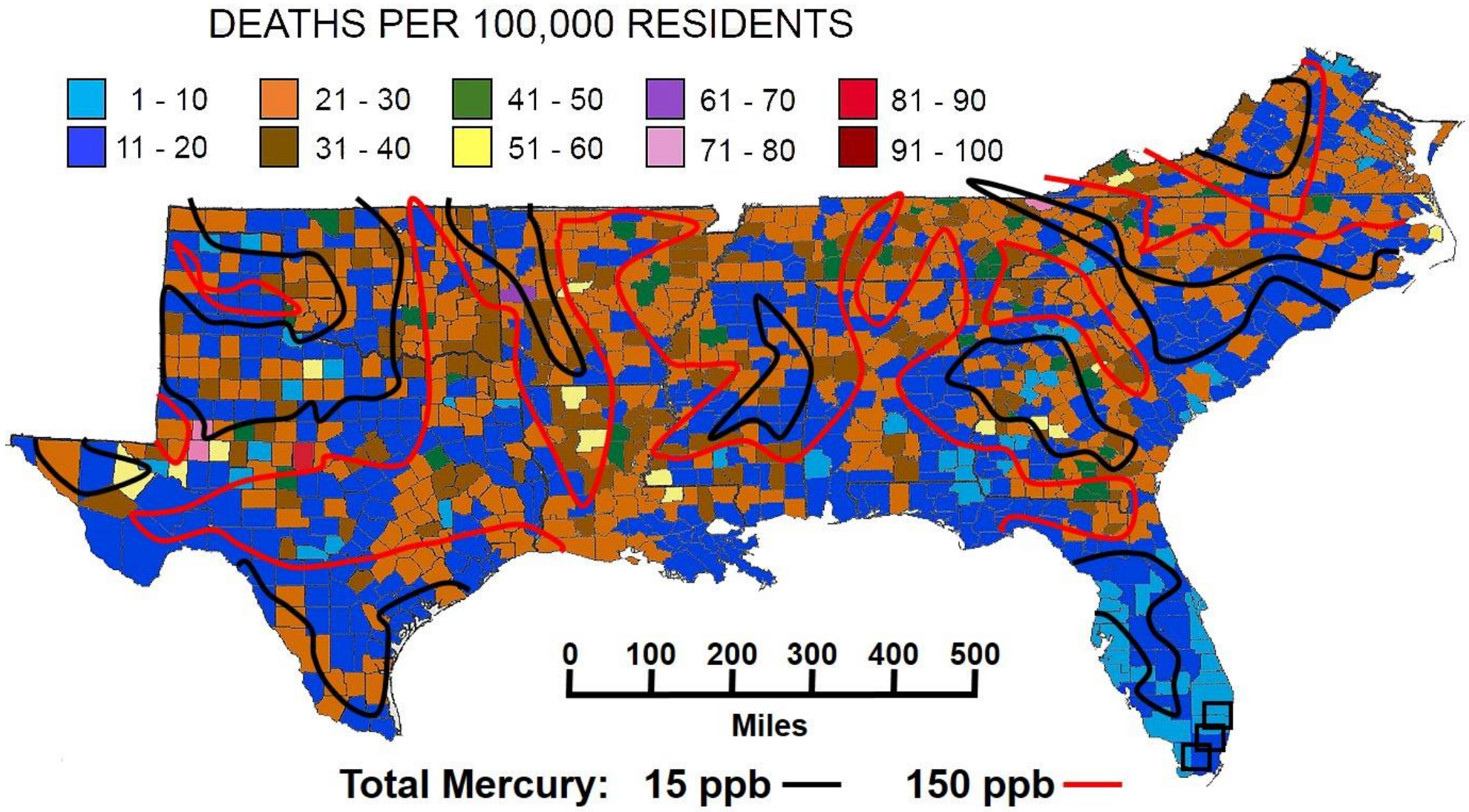

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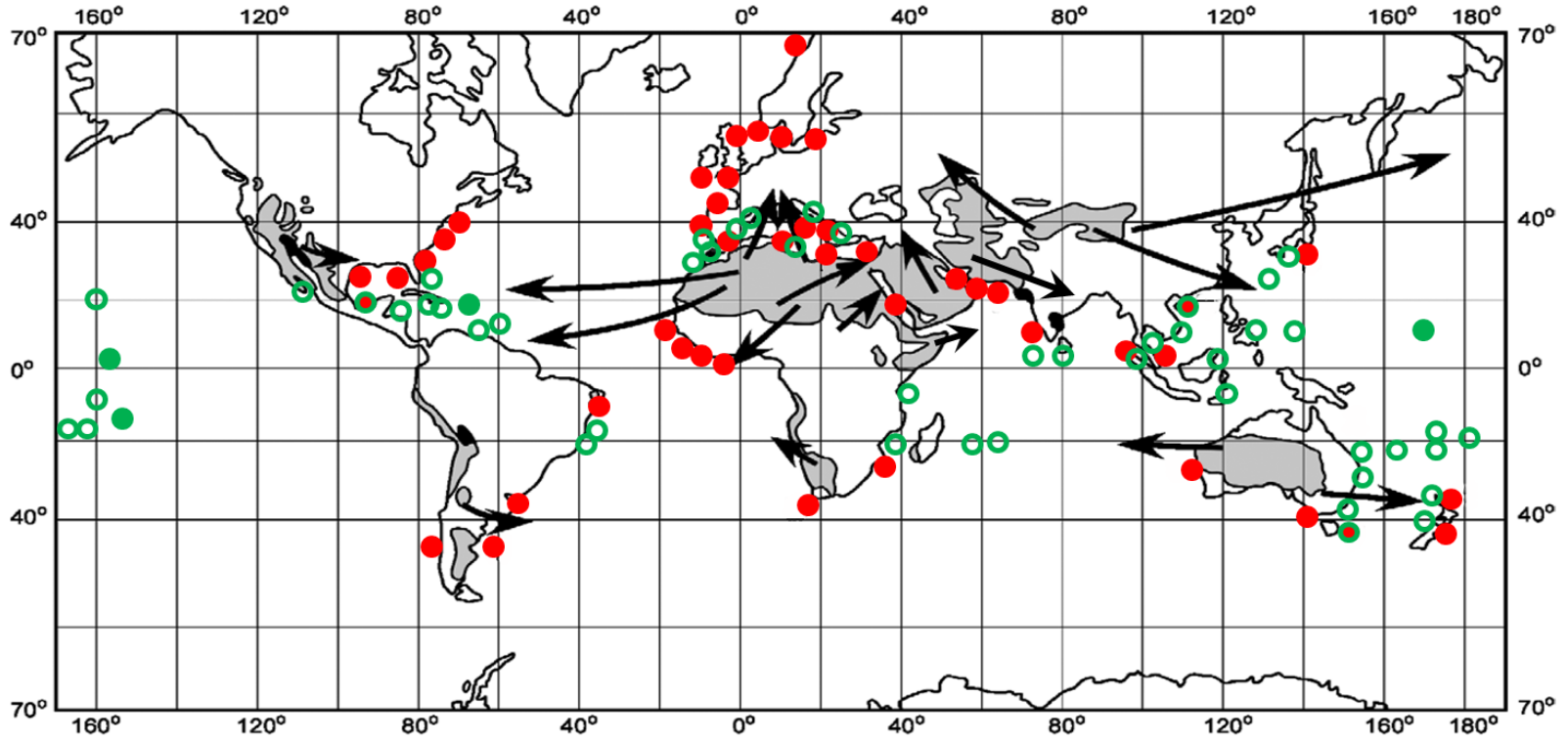
Global Connections Between Asthma & HABs Exacerbated By Human Activity



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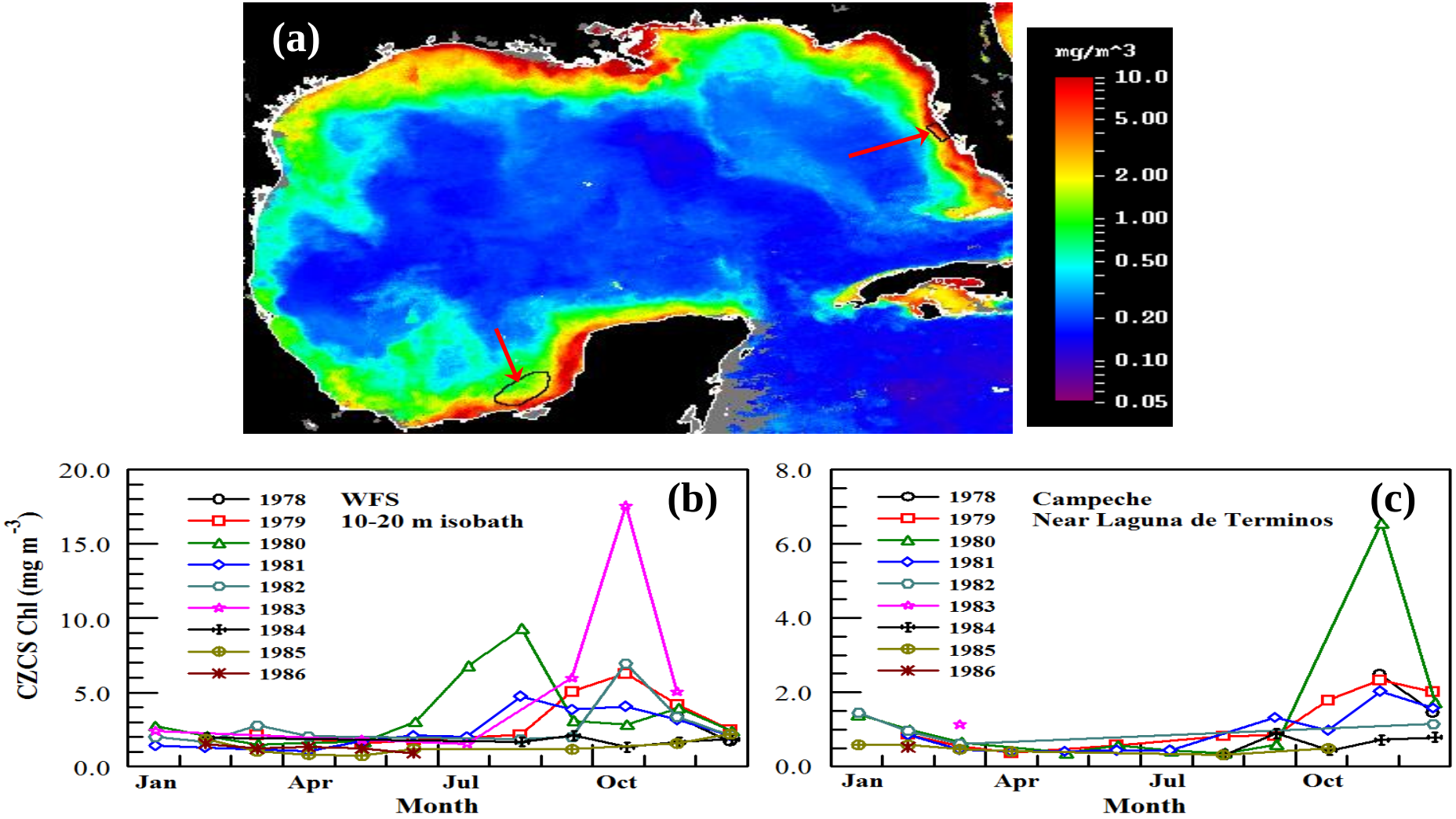

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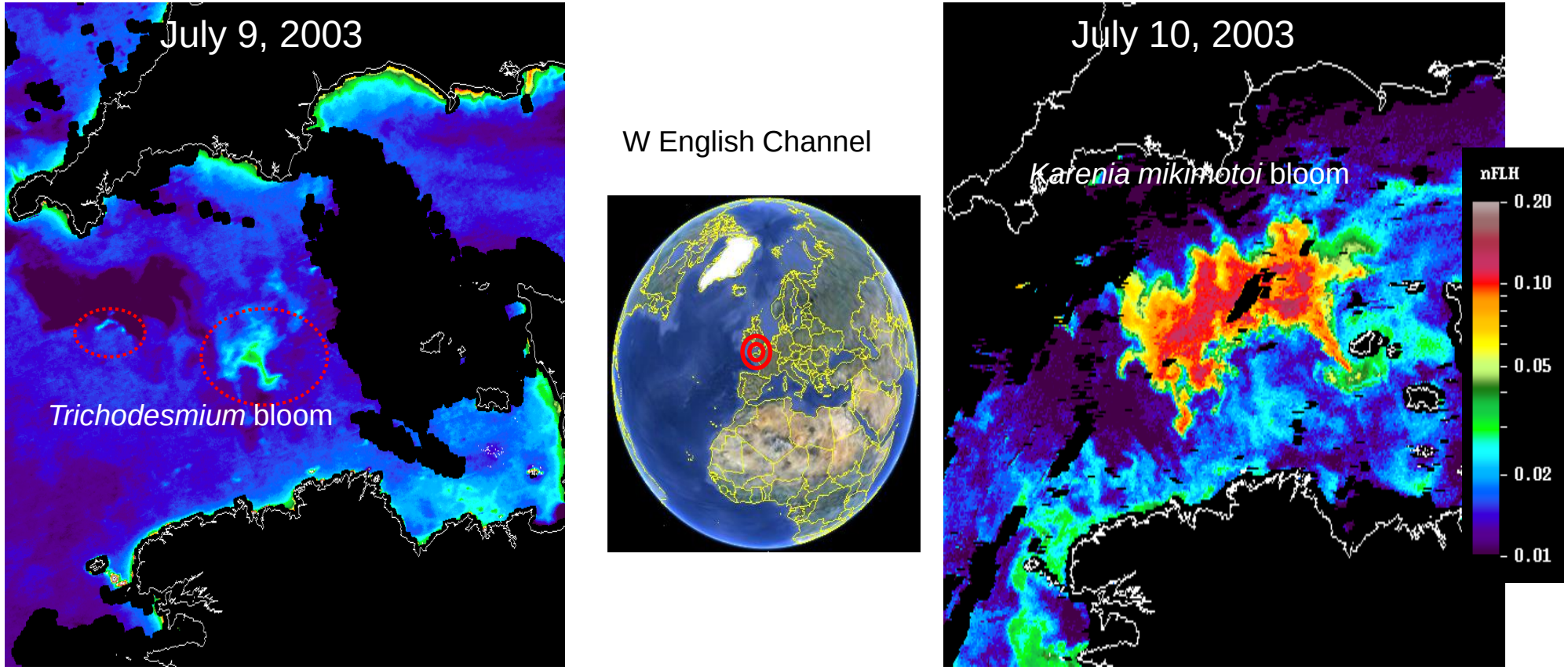

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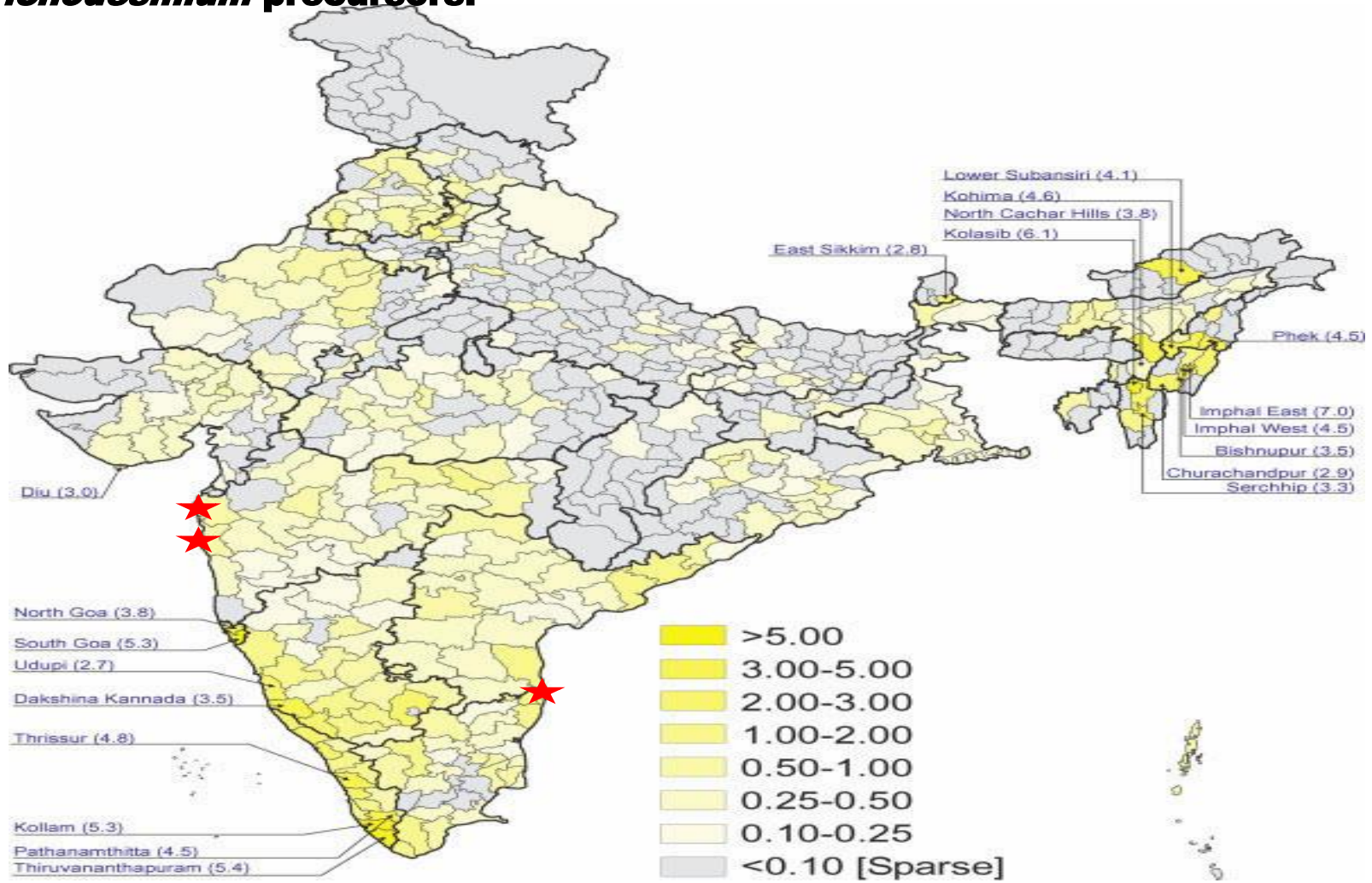

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