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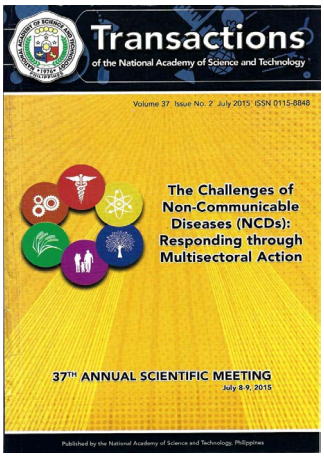

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CHEMICAL POLLUTANTS WITH NON COMMUNICABLE DISEASE HEALTH EFFECTS IN THE PHILIPPINE ENVIRONMENT

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Introduction

Key Non Communicable Diseases (NCDs) include autoimmune diseases, cancers, chronic cardiovascular diseases, chronic kidney diseases and chronic pulmonary diseases. NCDs are usually chronic which last for long periods of time and progress slowly. Among other factors like gender, genetics and lifestyle, exposure to environmental pollution has been identified as increasing the likelihood to develop NCDs.

Some pollutants have been determined to cause adverse health effects to humans and wild life. Their sources, introduction to the environment, pathways to human contamination and metabolic effects have been studied. In this paper, trace metals and organic pollutants that have NCD effects and which are deemed important in terms of their sources and usage in the Philippines are discussed. Some monitoring data in the Philippine environment will be presented. Some suggestions to address a number of existing chemical pollution in the country that can contribute to the development of more NCDs are presented.

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Trace Metals

Chemical and Biological Properties

Arsenic, cadmium, chromium, lead, mercury and nickel belong to the group of elements classified as transition metals. Transition metals have a positive charge in their natural state and possess electron orbitals that can be shared with electrons from other substances. In biological systems, arsenic, cadmium, chromium, lead, mercury and nickel can form covalent bonding with electrons from amino acids in proteins and enzymes; such interactions can result to dysfunctions of proteins and enzymes (Goyer R, Clarkson T, 2001).

Humans are exposed to trace metals from contamination of air, water and food. In air, metals are in the form of oxides and silicates in air particulates that can deposit in the lungs; inhalation of contaminated air is the primary way workers in mining and metal manufacturing are exposed. The general population is exposed to trace metals primarily through contaminated food. The toxicity of trace metals depends on their form, oxidation state and ease of absorption by the target organism. Water soluble arsenic and cadmium are easily absorbed by plants while more lipid soluble organic lead and organic mercury are more easily assimilated by fish, shellfish, animals and humans. Because of the similarity in the structure of phosphate (HPO_4^-), for which a natural mechanism for absorption exists in living things, the water soluble form arsenate (AsO_4H^-) of arsenic is more easily absorbed than arsenite (AsO_2H) and the water soluble chromate (CrO_4H^-) is easily more absorbed than chromite ($Cr_2O_4H_2$). However, the trivalent arsenic and trivalent chromium are more reactive and more toxic to biological materials. For metals that are not easily absorbed in the intestinal tract because of their large atomic weight such as lead and mercury, their methylated or organic forms which are lipid soluble allow these metals to pass the intestinal walls more easily. Once the metals are in the circulatory system, they react with blood proteins and are distributed in the body (Goyer R, Clarkson T, 2001).

Arsenic, cadmium, lead and mercury have high affinity for sulfur. In biological materials, the primary targets for interactions of these metals are the sulfur or thiol-containing amino acids such as cysteine, cystine, glutathione and the metallothionien proteins. Ligand binding to thiol groups in enzymes is the most common mechanism of toxic action of these trace metals (Goyer R, Clarkson T). Cellular targets are specific enzymes and/or receptors on membranes and organelles. Dysfunctions of enzymes in specific organelles are manifested in disorders in the organs that develop to noncommunicable diseases.

Lead, cadmium and nickel have electron charge densities similar to the essential metals calcium, iron and zinc respectively. The multiple toxicity of lead can be attributed to the displacement of calcium by lead in many of the metabolic functions of calcium in cell communication, energy activation and bone formation. Deficiency in iron induces the production of iron binding proteins and increases the absorption and toxicity of cadmium. Nickel has been shown to replace zinc in zinc binding sites of DNA-binding protein (Goyer R, Clarkson T, 2001). Table 1 shows the similarities in ionic radius and Vanderwaal's radius between the toxic metals and the essential metals.

Some metals such as cadmium, lead and nickel can form stable precipitates with molecular proteins that can damage the tissues. Cadmium, lead and mercury produce renal toxicity by clogging the renal tubules with metal-protein precipitates. Nickel is known to enhance the condensation of DNA binding proteins (Goyer R, Clarkson T, 2001).

The cellular reactions of arsenic, cadmium, chromium, lead, mercury and nickel that can lead to NCD diseases are shown in Table 2.

Sources of environmental pollution with trace metals

Mining of mineral ores is one of the major sources of pollution of toxic trace metals in the Philippine environment. In 2014, about 50 large scale mining projects and numerous small scale mining operations all over the country produced 42 kgms of gold and silver, 502 metric tons of concentrates of copper and nickel in ores and 867 dry metric tons of chromite and iron ores (MGB, 2014). Arsenic, cadmium, lead and mercury are extracted out of the earth together with the valuable iron, copper and gold in ores that are rich in sulfur. Mine workers are primarily exposed to arsenic, cadmium, lead, mercury and nickel in dust contaminated with these metals.

Extracting gold with mercury in many small scale mining sites is the most important source of mercury pollution in the Philippines (UNEP-DENR, 2010). Workers and their families living in these sites are directly exposed to mercury vapor while the surrounding land, water and fish resources are contaminated with inorganic and organic mercury. Population inside and outside of these gold extraction facilities are exposed to plant food produced in the surrounding areas and in fish caught in rivers and coastal areas in the vicinity of the facilities.

Volcanic eruption is the most important natural release of sulfur -rich minerals to the environment. The eruption of the long dormant Mt Pinatubo in 1990 released tons of sulfur dioxide and ash potentially containing arsenic, cadmium, lead and mercury to large areas in the provinces of Zambales, Pampanga and Tarlac. For many years during the rainy season, tons of lahar from the eruption had been washed away from rivers in these provinces to the coastal areas of Zambales. Among the other 18 active volcanoes in the country, Mayon in Albay, Taal in Batangas, Mt. Kanlaon in Negros Oriental and Mt Bulusan in Sorsogon erupted in recent times from 1996 to 2015.

The metals industry including steel and steel products manufacturing and scrap metal recycling are important sources of metals contamination of air and water effluents near industrial plants. A number of companies in the country produce nickel pig iron, and steel billets from recycled scraps. Metal scrap collection is usually done by the informal sector and the scraps are stored in open junkyards until these are sold to companies that can melt the scraps. Many local companies are producing metal products for construction such as reinforcing bars, nails, steel pipes and galvanized ion sheets. Other companies produce metal products as stainless steel kitchen materials, metal towers, furniture and jewelry.

Unsafe disposal of consumer products such as batteries containing lead, nickel and cadmium contributes to widespread contamination of soil and water with these metals. Continuous emission of combustion products of petroleum-based fuels in motor vehicles and in coal fueled power plants release toxic metals in the atmosphere and disperse the contamination of metals over wide areas.

Notable trace metals pollution in the Philippines

1. Pollution of the environment with mercury used in gold recovery in Mt. Diwalwal in Compostela Valley and in Paracale, Camarines Norte (UNEP-DENR, 2010).

Mercury concentrations of air, soil, fish, hair and blood of population in Mt. Diwalwal in Compostela Valley and in Paracale, Camarines Norte have been reported as exceeding standards for the protection of the environment and public health (Table 3). The data shows that fish sampled in Mt Diwalwal exceeded the safe concentration for human consumption. The mercury concentrations in ambient air and in the blowtorch area in Paracale showed unsafe conditions for residents and workers. In both Mt. Diwalwal and Paracale, the average blood concentrations in workers and residents sampled indicated that the people in these sites are at risk to mercury poisoning.

2. Pollution of the Meycauayan-Marilao-Obando (MMO) River System with toxic trace metals from various formal and informal industrial activities (ADB, 2009).

The MMO river system in Bulacan province originates from the mountainous areas of Angat, runs through the relatively highland areas of Marilao and Meycauyan and through the low lying areas of Obando before draining into the Manila Bay. The MMO river system has been the dumping ground of effluents that contain toxic metals and organic wastes from multitude of industries located along the river system. These industries include lead recycling, jewelry making, electroplating, tanneries, plastics and textiles. Other contributors to the pollution of the river system are the open dumpsite and piggeries in Marilao. The largest lead smelting company that recycles lead batteries is located in Marilao. Approximately 2000 jewelry makers and the associated industry of gold extraction are located in Meycauayan. Raw leather materials in making leather products in the country come from tanneries in Meycauayan. The waste coming from industries and piggeries in the watershed of the river system are dumped to the soil and river water without treatment. Baseline monitoring of the river system in 2008 showed that trace metal concentrations in many sampling sites exceeded the DENR EMB water quality standards for surface water and the Washington State standard for sediment. Tables 4a and 4b show the sampling sites with the highest concentrations of trace metals monitored in the river system.

The contamination of river water in Obando which receives the wastes from Marilao and Meycauayan is a major concern for public health because the water is distributed to the 888 hectares of freshwater and 15059 hectares of brackish fishponds in Obando, Bulacan, Paombong and Hagonoy that produce Bangus, Tilapia, oysters and other shellfishes. From the baseline concentrations reported, lead and mercury maybe available to aquatic organisms in aquaculture in Obando and in other neighboring towns. Table 4c lists the concentrations of toxic trace metals found in some biota.

The surface water and sediments in the river system are highly polluted with toxic trace metals. A limited sampling of biota in Obando showed that the shellfish Paros was unsafe for humans.

Monitoring of trace metals contamination in seafoods

The following are excerpts from several studies conducted on the concentrations of toxic trace metals in seafoods:

1. Mercury contamination of three fish species sold in Nepa Q Mart in Quezon City (Santiago E. et al, 2009)

The total mercury concentrations in bangus or milkfish (*Chanos chanos* Forskal), tilapia (*Oreochromis niloticus*), and galunggong or round scad (*Decapterus spp.*) purchased from Nepa Q Mart in Quezon City from 5 August to 20 October 2004 were determined by cold vapor atomic absorption spectrophotometry. The range of total mercury concentrations observed from about 30 composite test samples for each fish species were 0.0060 to 0.015 mg/kg (wet weight) for bangus, 0.0041 to 0.017 mg/kg (wet weight) for tilapia, and 0.014 to 0.05 mg/kg (wet weight) for galunggong.

Risk assessment for neurological effects associated with the consumption of the fish species with the highest concentration of mercury (0.05 mg/ kg for galunggong) was done. The calculated daily dose of total mercury of 0.06 μg $d^{-1} kg^{-1}$ body weight indicates that consumption of any one or any combination of bangus, tilapia, and galunggong sold in Nepa-Q-Mart from August 5 to October 20 in 2004 does not entail risk to adverse neurological effects.

2. Concentrations of trace metals in aquatic organisms from Albay Gulf after a mine tailings spill (Pascual A et al, 2008)

The different species of fish, small shrimps, shells and cuttlefish collected from selected localities in Sorsogon and Rapu-rapu island showed varying concentrations of mercury which ranged from less than the analytical method detection limit (MDL) of 0.02 mg/kg wet weight to 0.25 mg/kg wet weight. The highest concentration of mercury (0.25 mg/kg wet weight) among the fish samples was found in *Lutjanus decussatus* (Bangayaw, checkered snapper) caught in a large cove offshore Buenavista, Bacon, Sorsogon. The range of mercury concentrations detected in the other fish species (0.039-0.073 mg/kg wet weight) is higher than the range than the range (0.014-0.050 mg/kg wet weight) found in the pelagic marine fish *Decapterus macrosoma (galunggong)* sampled from a local market (Santiago et al, 2006). It is interesting to note that *Elagatis bipinnulata* (Bulangawan) a much bigger fish (56 cm) than *Lutjanus*

decussates (21 cm) had lower mercury concentration. Lead concentrations in the fish samples ranged from less than the MDL of 0.04 mg/kg wet weight to 0.21 mg/kg wet weight and the highest lead concentration (0.21 mg/kg wet wt) was found in *Selaroides leptolepis* (Atuloy) caught in the same large cove offshore Buenavista, Bacon, Sorsogon. The lead concentrations detected in the fish samples compare with the range of concentrations (less than MDL of 0.1 mg/kg wet weight to 0.23 mg/kg wet weight) found in pelagic fishes *Thunnus albacores* (yellowfin tuna), *Katsuwonus pelamis* (skipjack tuna) and *Thunnus obesus* (Bigeye tuna) in the survey conducted by the Bureau of Fisheries and Aquatic Resources of the Philippines (BFAR, 2001). Cadmium was not detected in most biota except in the flesh of *Cassis cornuta* (helmet shell) from the abandoned mine in Rapu-rapu island (0.03 mg/kg wet weight). Arsenic concentrations were higher in cuttlefish (15 mg/kg wet weight) and in the flesh of helmet shells (10.7 mg/kg wet weight) from the abandoned mine in Rapu-rapu than in small shells from a Sorsogon site (6.6 mg/kg wet weight). The mercury and lead concentrations in the fish investigated are below the internationally –accepted regulation limits of 0.5 mg/kg methyl mercury and 0.3 mg/kg lead (CODEX, 2006). The cadmium concentration in the helmet shells flesh from the abandoned mine in Rapu-rapu island is below the limit of 2 mg/kg for shellfish (CODEX, 2006). CODEX has no regulation limit for arsenic in fish, as the principal arsenic form in fish and crustaceans is arsenobetaine which is considered non toxic (CODEX, 2006).

3. Trace metal content in mussels, *Perna viridis L.*, obtained from selected seafood markets in a metropolitan city (Dumalagan H et al, 2010).

Mussels obtained from three selected seafood markets in Pasay and Paranaque, were tested for metal content including cadmium, copper, lead, and zinc through atomic absorption spectrophotometry. Among the trace metals analyzed in the soft tissues of mussels, only copper and lead exceeded the PEMSEA guideline for safe human consumption. The mussels sampled in this study showed an average lead concentration of 0.8 mg/kg and a maximum concentration of 2.3 mg/kg, which exceeded the PEMSEA guideline for lead (Table 5).

Suggestions for future studies

1. Monitoring and risk assessment of cadmium, chromium, lead, mercury and nickel contamination of fish and raised in polluted areas like Obando, Meycauayan and Marilao in Bulacan and in the coastal areas of Mt. Diwalwal in Compostela Valley.

2. Monitoring and risk assessment of the potential contamination of cadmium and arsenic in rice grown in areas where volcanic eruption had occured.

3. The potential risk to adverse effects of arsenic, cadmium, chromium, lead, mercury and nickel from inhalation of 2.5 micron air particulates in EDSA and in some residential areas in the Greater Manila Area.

Organic Pollutants

The chemical organic pollutants that could cause adverse health effects to biological systems have molecular structures that are not easily degraded in the environment and in biological systems. These pollutants are usually water insoluble and lipid soluble. These characteristics enable the pollutants to pass the cell membrane of living organisms and react with biological molecules inside the cell. Some toxic organic pollutants can substitute for naturally occurring biological compounds with similar structures and express the function of these endogenous materials. Other toxic organic pollutants interact with proteins and enzymes and cause dysfunction of these biological substances in the living organism (Capen C, 2001). The group of persistent organic pollutants (POPs) as pesticides, as industrial chemicals Polychlorinated Biphenyls (PCBs) and Polybrominated diphenyl ethers (PBDEs) and as Dioxins and Furans, the class of Polycyclic aromatic hydrocarbons (PAHs) and some alkyl phenols are some of the ubiquitous organic pollutants in the Philippine environment that can cause NCD health effects. These organic pollutants can disrupt the functions of hormones and enzymes that can result to chronic diseases, reproductive defects and cancers.

Chemical and Biological Properties

1. Persistent Organic Pollutants (POPs)

POPs are compounds that consist of aromatic rings with multiple chlorine or bromine atoms. These compounds are resistant to chemical degradation by hydrolysis or by oxidation –reduction in the environment. POPs are semi-volatile, present in air as vapor or adsorbed in dust particles and can be transported by air to distances away from their origin. POPs deposit in water bodies by runoff of contaminated soil or by wet and dry deposition of contaminated air. Humans are exposed to POPs through the food chain and through inhalation of contaminated air. Occupational workers are exposed to POPs through the skin and respiratory system.

a. POPs Pesticides

Aldrin, endrin, dieldrin, heptachlor and in lesser extent DDT were the popular pesticides used in rice, vegetable and fruit farming in the country in the 1960's up to early 1980's. Other chlorinated pesticides were used mainly for different purposes; DDT for control of malaria infestations in forested areas and chlordane for the control of termites in residences in the urban areas.

The pesticides DDT, dieldrin, methoxychlor, chlordane and lindane are associated with endocrine disruption, carcinogenicity, immune suppression and neurotoxicity (Table 6).

b. POPs Industrial Chemicals Polychlorinated biphenyls (PCBs) and Polybrominated diphenyl ethers (PBDEs) PCBs and PBDEs are both halogenated aromatic chemicals commercially manufactured for different purposes.

Commercial transformer oils containing PCBs were widely used in electric distribution in the country for many years before 1980. The indiscriminate handling and disposal of PCB oils in some electric distribution facilities and transformer repair facilities is the major contributor of PCB contamination of soil. In 2004, an administrative order from the Department of Natural Resources and Environment placed PCBs under a chemical control order which aims to eliminate the existence of PCBs in the country (DENR, 2006). The chemical control order for PCBs requires all users of PCBs to register with the Environmental Management Bureau (EMB) and to report the safe management and disposal of PCBs.

PCBs are a class of synthetic compounds characterized by two phenyl rings with chlorines attached to the phenyl rings (Table7). PCBs are formed by chlorination of biphenyl and the process can produce up to 209 congeners (compounds with the same structure but differing in the number and placement of chlorine in the ring). The chemical and physical properties of the PCB congeners differ according to mass, chlorine content and position of chlorine in the biphenyl rings. The water solubility decreases with higher chlorine content, the organic matter partitioning (log Kow) increases with higher chlorine content. The toxicity of PCBs increases with the number of chlorines in the biphenyl ring. PCBs are stable at high temperature and have a high dielectric constant, characteristics that make PCBs ideal as a heat transfer fluid.

PCBs are concentrated in food, ingesting food is the most important pathway of exposure to PCBs for ordinary people. Skin absorption and inhalation are more important pathways for occupational workers' exposure. PCBs can cross the gastrointestinal tract very rapidly because they dissolve very easily in the lipid linings of the intestines. PCBs accumulate in the liver since going to the liver is the direct path of absorbed substances from the intestines. Once carried in the blood stream, PCBs can deposit to adipose tissues. The high affinity of PCBs to lipids enables PCBs to cross the fatty layer of placental barrier that protects the fetus from contaminants. PCBs with higher chlorine content are less accessible to enzymes that can metabolize the PCBs, thus they stay in the body longer to cause more toxic effects (Capen C, 2001).

c. POPs Polychlorinated Dioxins (PCDDs) and Furans (PCDFs)

Polychlorinated dibenzo dioxins (also known as Dioxins) and Polychlorinated dibenzo furans (also known as Furans) are persistent halogenated aromatic compounds (Table8). There are 75 congeners of PCDDs, seven of them are specially toxic with the 2,3,7,8-tetrachlorodibenzo-*p*-dioxin (TCDD) as the most toxic. There are 135 congeners of PCDFs and ten of them are toxic. Dioxins and Furans are not made intentionally but they can be produced as by-products of manufacturing of other chemicals such as the herbicide 2,4,5-Trichlorophenoxyacetic acid and the fungicide pentachlorophenol. Dioxins and Furans can also be produced during the process of bleaching of pulp paper and wood. Burning of organic matter especially plastics and chloride-containing materials produce Dioxins and Furans (USEPA, 2002). Uncontrolled combustion processes was found to be the biggest source to releases of Dioxins and Furans to air, land, and water (USEPA, 2002). The inventory of Dioxins and Furans (DENR, 2006) indicated high releases from uncontrolled household and agricultural burning, open dumpsites and landfill sites. All open dumpsites and landfill sites in the country are potential contaminated sites for Dioxins and Furans. Dioxins and Furans are released in the air and transported in the atmosphere. The general population is exposed primarily to contaminated food; scavengers in dumpsites are exposed to contaminated air. The country has no existing analytical capability to monitor Dioxins and Furans and has not validated the high releases of these toxic compounds in the environment.

Toxic effects of POPs

PCDDs and PCDFs like the PCBs and PBDEs cause toxicity through the activation of the aryl hydrocarbon receptor (AHR). TCDD has been implicated in genetically inherited susceptibility to breast cancer by epigenetic mechanism through methylation of DNA proteins of the breast cancer tumor suppression gene (Hockings et al, 2006)

Halogenated aromatic POPs cause toxicity primarily by inducing the dysregulation of gene expression. PCBs, PBDEs, PCDDs and PCDFs act as stable ligands of aryl hydrocarbon receptor (AHR). The interaction of the activated AHR with transcription factors of many genes could result to either enhancement or inhibition of the expression of the gene's function. POPs induce production of the enzyme cytochrome P450, a monooxidase enzyme that promotes the production of oxidizing xenobiotics that initiate cancer. Through the activation of AHR, POPs have been found to interact with other genes expressing metabolic enzymes that result to various metabolic defects. POPs have been implicated in promoting cancer by dysregulation of the *ras* gene and tumor suppressor gene (Capen C, 2001). Table 7 shows some of the cellular effects and toxic effects of PCBs PBDEs, Dioxins and Furans.

d. Polycyclic Aromatic Hydrocarbons (PAHs)

PAHs are a group of chemicals consisting of carbon and hydrogen in multiple fused aromatic rings. Most PAHs are produced from incomplete combustion of plant or animal matter, or carbon fuels, such as coal or petroleum. Natural (forest fires) or man-made practices of burning produce PAHs. Automobile exhaust, industrial emissions and smoke from burning wood, charcoal and tobacco contain high levels of PAHs (NAP, 1985). In general, more PAHs form when organic materials burn at low temperatures, such as in wood fires or cigarettes. High-temperature furnaces produce fewer PAHs. Smoke or emissions contain PAHs in the form of air particulate or in gaseous form. Generally, PAHs with higher number of rings (>5rings) adsorb to air particulates and PAHs with lower number of rings (<5) partition to the gas phase. PAHs have great affinity for lipids; when released in the environment, PAHs adsorb on organic matter of dust particles and soil which eventually become part of the surface sediments of waterways, estuaries, bays and open sea (Valls et al, 1991). Humans are exposed to PAHs through the food chain as well as through contaminated air. In Metro Manila, with its dense population, heavy industrial activities and uncontrolled vehicular emissions, the contamination of the environment with PAHs is expected to be significant.

PAHs vary in the number of aromatic rings and in the manner the rings are fused together. The stability of the PAHs depends on the size and the type of fusion of the rings, hence the chemical and biological reactivities of PAHs are different (USEPA, 1984). Unlike POPs, PAHs in the environment can be degraded by photooxidation to peroxides (Khan et al, 1967) or by microbial enzymes of aerobic bacteria and fungi (Thomas et al, 1986). In biological systems, PAHs react with DNA and other macromolecular nucleophiles after transformation into epoxides or phenols either by photochemical oxidation or by metabolic activation with mixed function oxidases. The final opening of the epoxides to diol presents a positive center which reacts with macromolecular nucleophiles like DNA (Zanders, 1983). The structure of the PAH is important to the covalent bonding of PAH with DNA. The ability of the PAH to sterically hinder the formation of diol epoxides in its structure and favor the formation of a stable carbonium ion has been associated with carcinogenicity of some PAHs (Pullman and Pullman, 1955). PAHs proven to have strong carcinogenicity to mammals are 7,12 dimethylbenz(a)antracene, dibenz(a,j)anthracene, benzo(c)phenanthrene, benzo(b)fluoranthene, benzo(j)fluoranthene, cholanthrene, 3-methylcholanthrene, benzo(a)pyrene,

dibenzo(a,h)pyrene, and dibenzo(a,i)pyrene (USEPA, 1984). The carcinogenicity of PAHs is attributed to 1) the induction of point mutation in the *Ras* gene and 2) induction of Cytochrome *CYP1A1* as tumor promoter by altering the gene expression of tumor suppressor gene and other xenobiotic metabolizing enzymes. The cellular and toxic effects of representative carcinogenic PAHs are listed in Table 8.

e. Phenols

Phenols are used as chemicals in detergents, lubricants, coatings, polymers, resins, antioxidants, and pesticides among other industrial products. The alkylphenol nonylphenol, bisphenol A (BPA) and pentachlorophenol (PCP) are some of the phenols widely used in food manufacturing, wood processing, textile and leather manufacturing and are known as endocrine disrupting chemicals (EDCs). EDCs are known to interfere with the hormonal systems, affecting growth, development and reproduction in humans and animals (Colburn et al., 1993). Animal and human studies have shown that nonylphenol and BPA cause reproductive impairments by affecting the functions of estrogen. EDCs with estrogenic effect have similarity in structure with the endogenous estrogen, such that they can substitute for estrogen as ligands in the estrogen receptor site. On the other hand, PCPs affect the functions of thyroid hormones and thymus cells that result to neurotoxicity and immune function defects by interefering with the regulation of the required levels of immune cells and thyroid hormones to preserve the balance in the body. Table 9 shows the cellular effects and toxic effects of nonylphenol, bisphenol A and pentachlorophenol.

Nonylphenol are components of industrial surfactants and detergents, and are additives to hundreds of industrial and consumer products for carpet and dry cleaning, pesticides, paper manufacturing, paints and coatings. Common sources of exposure to nonylphenol for the general population are cosmetics, household cleaners and paints. Nonylphenol is bioaccumulated by aquatic organisms and humans may be exposed to nonylphenol through consumption of contaminated fish and shellfish. Nonylphenol has been shown to have estrogenic properties in human cell lines and laboratory animals. Human cells surrounding the fetus at very early gestation may be highly responsive to the estrogenic effects of nonylphenol (Capen C, 2001).

BPA is a chemical used in the manufacture of polycarbonate plastics and epoxy resins such as food and drink containers, plastic water bottles, baby bottles, dental sealants and a variety of household products. Human beings are mainly exposed to BPA via dietary ingestion of leachings from the inner lining of cans and microwave containers during heating of food materials or from polycarbonate beverage containers from repeated usage or contact with acids and alkali. In humans, BPA has been found to have estrogenic properties affecting the regulation of hormonal function of both fetal and adult male hormones (Capen C, 2001).

PCP is a chlorinated phenol used as herbicide, insecticide, fungicide, algaecide, disinfectant and as an ingredient in anti-fouling paint. PCP is applied as preservative in leather, masonry, rope, and wood and as disinfectant in cooling tower water and paper mills. Workers in facilities using PCPs may be exposed through inhalation of contaminated air or through skin exposure. The general population may be exposed by dermal contact to treated wood or by ingestion of contaminated water and food. Occupational exposure to PCPs has been associated with non-Hodgkin's lymphoma and multiple myeloma (Table 8).

Monitoring of organic pollutants in the Philippine environment

1. Endocrine-disrupting phenols in selected rivers and bays in the Philippines (Santiago and Kwan, 2007)

Endocrine disrupting phenols were surveyed in surface and bottom waters in river and coastal sites in Manila Bay, Subic Bay and Batangas Bay for two years in 2000-2002. Among the study sites, the rivers in the Greater Manila Area (GMA) showed the highest average concentrations for most of the detected phenolic compounds.

Meycauayan River, Tenejeros River and Parañaque River in the GMA showed concentrations of 4-n-nonylphenol at 79 μg/L, 4.5 μg/L and 5.0 μg/L respectively. Pasig River showed the lowest average concentration of 4-n nonylphenol at 0.46 μg/L and the highest concentration of 2,4, dichlorophenol at 0.19 μg/L. The levels of pentachlorophenol at 0.91 μg/L and bisphenol A at 0.80μg/L were highest in Meycauayan and Tenejeros Rivers respectively. All rivers outside GMA and all the bay sites including Manila Bay showed phenols in varying average concentrations below 1 μg/L. The data showed that the Meycuayan river and Tenejeros river are highly polluted with endocrine disrupting compounds which are drained to the Manila Bay at the mouth of Obando river.

POPs pesticides

a. POPs pesticides in selected rivers and bays in the Philippines (Santiago E and Kwan C, 2015)

POPs pesticides were monitored for three years in 2002 -2005 in water samples from rivers in Pangasinan and river tributaries of Laguna Lake to determine the types and concentrations of organochlorine pesticides that may contaminate the two important water resources for aquaculture in Luzon. Traces of hexachlorobenzene (BHC), aldrin, trans and *cis*-chlordane, dieldrin, DDT and metabolites, nonachlor, endosulfan and endrin ketone in water samples from both provinces were detected at levels near the analytical method detection limits (MDLs). During the first year of monitoring, the highest concentrations of Total OCPs (sum of the average concentrations of OCPs detected above the MDLs in water samples were detected during the rainy season in September 2002 in Laguna at 0.25μg/L and during the dry season in January 2003 in Pangasinan at 0.35μg/L. On the third year of monitoring, the highest Total OCPs concentration in Laguna was 0.03μg/L in Dec 2004 and 0.08μg/L in Pangasinan in January 2005. The results showed that the organochlorine pesticide residues in the watershed areas of the two river systems are decreasing with time.

POPs pesticides were monitored in 2004-2005 in sediments from the same sampling sites where water samples were collected in Pangasinan and Laguna Lake river tributaries. In Laguna and Rizal *trans*- and *cis*-chlordanes, α-BHC, *p,p'*-DDT and its metabolites, endrin ketone, endrin aldehyde, endosulfan 2, and endosulfan1 with a total OCP concentration of 12.5 ng/g dry wt were detected (Fig 1a). During the second sampling the same OCPs were detected but gave a less total OCPs concentration of 10 ng/g dry wt. In Pangasinan, during the first sampling only *p,p'*-DDT and α-BHC were detected in sediments, the total OCPs concentration detected in the river system is less than 2 ng/g dry wt. During the second sampling higher levels of *trans*- and *cis*- chlordanes were detected in all the sediments from the seven river sites. Endosulfan1, endosulfan 2 and endrin ketone were detected in six of the seven sites. The metabolites *o, p'*-DDT, *p,p'*-DDD, *o,p'*-DDD and *p,p'*-DDE were detected in four sites. The total OCPs concentration detected in Pangasinan river system is 23 ng/g dry wt (Fig1 b). In both river systems, chlordane, DDT and its metabolites, endosulfans, and endrin ketone are more predominant in the sediments than other types of OCPs.

An alternative way of measuring environmental contaminants is measuring their concentrations in animals that are directly exposed to these chemicals through their ambient environment (Phillips, 1995). Freshwater shrimps, fish and squid were monitored in one river system and several coastal waters to assess the pollution of organochlorine pesticides in the natural waters. Shrimps *(Macrobrachium rosenbergii rosenbergii* and *Macrobachium rosenbergii daquete) c*ollected from two sites Pampanga river and in a river in Bay, Laguna in 2006 showed residues of *trans*-chlordane (1.98 - 2.55 ng/g wet wt) and metabolites of DDT(2.55 - 5.40 ng/g wet wt) in the muscle tissues. Among the samples of fish (*Lates Calcarifer*) collected from three coastal sites in Southern Luzon, only the fish caught in a coastal site in Tagkawayan, Quezon showed contamination of organochlorine pesticides with *trans*-chlordane (1.02-1.86 ng/g wet wt) and BHCs (3.0-9.5 ng/g wet wt.) detected in the fish muscle. OCPs were detected in liver and muscle tissues in all the squid samples collected from La Union Bay, Manila Bay, Tayabas Bay , Lamon bay and Visayan Bay during the dry season. All liver and muscle samples showed high contamination of β-BHC. In addition to β-BHC, other OCPs were detected in squid samples depending on the location of the sampling site. In the liver and muscles of squid, *o,p'*DDT, δ- and γ-BHCs were detected in the La Union , HCB, *trans*-nonachlor, δ-BHC, methoxychlor in Manila Bay ; γ-BHC, *trans*-nonachlor, *p,p'*-DDT, and *o,p'*-DDT in Tayabas

Bay; δ-BHC, *p,p'*-DDD and methoxychlor in Lamon Bay and γ-BHC, endosulfan and methoxychlor in Visayan Bay. β-BHC was detected in most liver and muscle samples collected during the rainy season. The liver samples from Manila Bay showed the most number of OCPs detected; including *cis*-chlordane, dieldrin, endrin, *cis*- nonachlor, *trans*-nonachlor and methoxychlor. Only β-BHC was detected in all the squid muscle samples collected from all the sampling sites except in Lamon Bay where *o,p'*-DDD and in Visayan Bay where *trans*-chlordane were also detected.

a. Organochlorine pesticides in air in selected urban and rural residential areas by passive sampling with polyurethane disk (Santiago E and Cayetano M, 2011)

POPs pesticides were monitored simultaneously in ambient air three urban and three residential areas for four sampling periods at different weather conditions in 2005 using passive samplers with polyurethane disks (PUF). Generally, chlordanes (α and γ chlordane and cis and trans nonachlors), endosulfans (endosulfan I, endosulfan II and endosulfan sulfate) and endrins (aldrin, endrin and dieldrin) were the major contributors to the OCP contamination in air in all the sampling sites. The group of DDTs (DDTs, DDEs and DDDs) gave the lowest concentrations among the OCPs monitored. The total OCPs sequestered on the PUF disk (sum of concentrations of detected OCPs) in all the sites ranged from 33-398 ng/disk, with the highest concentrations found in most urban sites. Using the sampling rate 4.59 m^3/day, which was derived from depuration experiments in the study, the concentrations of the pesticides in air were calculated. Among the urban sites, Parañaque consistently showed highest concentrations of OCPs in all the sampling periods. Chlordanes (sum of concentrations of α and γ chlordane and cis and trans nonachlors) were found predominant in the urban areas in all the sampling periods, while endosulfans (sum of concentrations of endosulfan I, endosulfan II and endosulfan sulfate) and endrins (sum of concentrations of aldrin, endrin and dieldrin) were observed in different concentrations at different sampling periods in the study sites. Chlordanes, which could be attributed to its use as termiticides in the urban residential villages, were found in the ambient air in the urban sites consistently in relatively higher concentrations (218- 2324 pg/m^3) than endrin (ND- 693 pg/m^3) and endosulfans (ND-461 pg/m^3) during the four periods of investigation. Endosulfans were detected in the farming area in Balagtas,Bulacan in all the sampling periods (102 -904 pg/m^3) while endrins, principally dieldrin was

detected (61-234 pg/m^3) in Rizal. Fig 2 shows the average total concentrations of organochlorine pesticides detected for the four sampling periods. The pollution of chlordane in air was confirmed in the results of a global study of POPs in air where the air sample collected from an urban area in Manila in 2005 gave the highest concentration among the samples in the study (Shunthirasengham C et al, 2010).

1. Polychlorinated biphenyls in selected tributaries of Pasig River and Laguna Lake (Santiago E and Rivas F, 2012)

During the first sampling in August, 2010, the total PCB concentration (sum of concentrations of PCB congeners detected from mono to deca PCBs) in Pasig river is highest in San Juan tributary at Sanchez Bridge (12.8 ng/L). This implies that the vast land area in GMA covering the creeks that drain to San Juan River is the main source of PCB pollution in Pasig River. These areas are Quezon City and San Juan City covering an area of 166 km^2. The lowest total PCBs concentration (0.5ng/L) was found in the most upstream sampling site in Pasig River in Marikina while the most downstream site at Jones Bridge showed the next highest total PCBs concentration. The PCB concentration (2.65ng/L) in Manila Bay is lower than the site at Jones Bridge which could be attributed to dilution effect in the estuary. The total PCB concentrations in water samples from selected sites in Laguna Lake in Taguig (0.3 ng/L), in Sucat (0.1 ng/L) were much lower compared to the sites in Pasig River. PCBs were not detected in in Sta Rosa, Laguna.

The water samples, collected in November after the big flood event caused by the typhoon Ondoy in GMA, generally showed higher contamination of PCBs. The sampling sites in Manila Bay and in Marikina showed more than twice the contamination observed during the first sampling. The site at Jones Bridge showed much lower total PCBs (1.2 ng/L); which could be attributed to the flushing effect of the high current of water towards Manila Bay. The site at Sanchez Bridge in San Juan still showed the highest concentration on second sampling. During the flooding, excess flood waters from Pasig River and from areas around Laguna Lake filled the lake beyond its maximum capacity. During the second sampling in November, after nearly two months of the flooding event in September 26, 2009, most communities around the lake were still flooded. The total PCB concentrations measured at Taguig (2.2 ng/L), Sucat (5.6 ng/L) and Sta Rosa (1.3 ng/L) were significantly higher during the November sampling (p<0.05) than the

concentrations obtained during the first sampling by t test for significance of the two mean concentrations.

The study showed that the soil and sediments in the watershed of Pasig River and Laguna Lake are contaminated with PCBs, and some PCBs are released to the water during a flooding event.

2. PBDEs from leachates in Payatas dump in Quezon City (Kwan et al, 2013)

Polybrominated diphenyl ethers (PBDEs) are extensively used as flame retardants in many consumer products. In the absence of a separate disposal facility for electronic wastes and other products treated with PBDEs, these consumer products are dumped in municipal disposal facilities. In this study, the concentration and nature of PBDEs in leachates from municipal solid waste dumping sites (MSWDS) of eight tropical Asian countries including the Payatas in the Philippines were determined; and the propensity of MSWDS as sources of PBDEs in aquatic environment was evaluated. Particulate matter from leachates collected in Payatas landfill and sediments from several points in Pasig river were analyzed for PBDEs to determine the concentrations and the PBDE congeners present in the samples. Fig. 4 shows that tri BDEs (BDE 17) up to deca BDEs (BDE 209) are found in the leachates; indicating that products containing commercial pentaPBDE, commercial octaPBDE and commercial decaPBDE were dumped into the Payatas landfill. The concentrations of the tri BDE to hepta BDE in the sediments are much lower than the corresponding concentrations in the leachates, however, the profile of PBDE contamination found in the sediments from Pasig river is similar to the profile of PBDE contamination in the leachates from Payatas landfill. The result of the investigation showed that leachates from Payatas could be important sources of toxic PBDEs in the aquatic environment of Pasig river.

3. PAHs

a. Levels of polycyclic aromatic hydrocarbons in PM_{10} air particulates from selected sites in the Greater Manila environment (Santiago, E, 2002)

The PM_{10} particulates were collected about 3-5 m from the roadside in four sites (North EDSA, Cubao, Makati and Pasay) along the stretch of Epifanio de los Santos Avenue (EDSA), in two industrial sites (Paranaque and Valenzuela) and in three residential areas (PhilAm Subdivision in Quezon City, Santiago Subdivision in Valenzuela, Sun Valley Subdivision in Paranaque) within the Greater Manila Area during two sampling periods in 2000- 2001. The average of the combined concentrations of the 23 PAHs determined for the two sampling periods ranged at 5.06 ng/m^3 to 124.1 ng/ m^3. The combined concentrations of carcinogenic PAHs (7,12 dimethylbenzanthracene, dibenz(a,h)anthracene, benzo(b)fluoranthene, benzo(k)fluoranthene, benzo(a)pyrene, indeno[1,2,3-cd] pyrene and benz(a)anthracene) comprised 55-67% of the total concentration of the PAHs in PM_{10} particulates. Benzo (a) pyrene was detected in almost all the sites at concentrations above 1ng/m^3, the World Health Organization reference concentration in air for cancer risk of 10^{-4}. Principal component analysis (PCA) and SAS Method Varimax Factor analysis on the locations and the concentrations of PAHs indicate that the industrial site in Valenzuela, the site at Cubao in EDSA, the industrial site in Paranaque and the site at Makati in EDSA as a group with highest PAH loadings in air particulates and the residential sites in Valenzuela and in Paranaque as a group with the least PAH contamination. The PAH contamination in North EDSA and in Pasay EDSA and in the residential site at Phil Am is in a concentration range between those of the two groups.

a. Polycyclic Aromatic Hydrocarbons in ambient air in selected urban and rural residential sites (Santiago E, Cayetano M, 2007)

The PAHs in ambient air in three urban residential sites (PhilAm Subdivision in Quezon City, Santiago Subdivision in Valenzuela and Sun Valley Subdivision in Paranaque) and in three rural residential sites (Taytay, Rizal, Sta Rosa, Laguna and Balagtas, Bulacan) were investigated by deploying a passive sampler with a polyurethane foam disk for six weeks and analysis of the PAHs adsorbed on the foam disk with gas chromatography /mass spectrometry. Air samples were obtained from the six sampling sites simultaneously at different weather conditions in 2005. About 99% of the PAHs detected by the method were PAHs with less than 5 aromatic rings. Among the PAHs with more than 5 rings that were detected were the carcinogenic benzo(b)fluoranthene, benzo(b)fluoranthene, benzo(a)anthracene and benzo(a) pyrene. The concentration of PAHs in ambient air is primarily affected by the conditions of temperature, humidity and proximity to sources of PAHs. Fig.5 shows that the concentrations of PAHs in the urban sites in Valenzuela and Quezon City are higher than the other urban site in Paranaque and the PAHs concentration in the rural sites in Laguna and Rizal are higher than in the site in Bulacan. The patterns of the concentrations of PAHs against the sampling time are the same for all the urban and rural sites since all the six sampling sites had more or less the same weather conditions at the time of the study. The PAHs in ambient air were lowest in the second sampling when several storms hit the country during this period of sampling and cleared the air of the pollutants. The concentrations of PAHs were highest in the fourth sampling when the temperature was cooler and humidity was less. The investigation showed that PAHs are present in ambient air of residences in the Greater Manila Area and in the neighboring provinces and the residents are potentially exposed to inhalable PAHs and to some carcinogenic PAHs all the time.

Implications of management of chemical pollutants to the prevalence of NCD in the country

Advances in toxicology provide us a better understanding of the cellular effects and toxic effects of chemical pollutants on the exposed individuals. The presence of different types of chemical pollutants in the Philippine environment with their varied target metabolic dysfunctions in the body, makes exposure to chemical pollutants an important risk factor in the development of different types of chronic diseases in the country. The knowledge on the polluted areas, the effect of the pollution in the immediate environment and the potential extent of harm to people outside its immediate environment are information necessary to protect the general population from the effects of the chemical pollution. In addition, the knowledge on the important pathways where the general population maybe exposed to the pollutant, the number of people that maybe affected by the pollution and actual monitoring of the pollutant in these pathways are important tools to prevent the risk of widespread development of diseases associated with the chemical pollutant.

The mercury pollution in Mt Diwalwal and in Paracale have been unabated for many years; and the people living in these communities have been determined to be at risk for mercury poisoning a long time ago. Repeated interventions by the government to convince the miners to shift to a method of gold extraction without using mercury have failed because of economic reasons, however, more serious efforts by the mining, health and economic regulatory bodies should be done to stop the practice. The potential debilitating neurological effects and mental retardation of adults and children and future children in the communities would seriously affect the quality of life of thousands of Filipinos. The mercury pollution and the risk of exposure will continue to persist until the miners are convinced that using mercury for gold extraction is more disastrous than beneficial to them. Population studies in the communities that would show how many miners and members of their families have neurological disorder and how many children are suffering from mental disorder, have not been done in spite of the long standing health risk of mercury exposure. Perhaps results of such study presented to the communities may help the miners realize the disastrous effects of mercury in their community and perhaps make the government take more serious actions to protect Filipinos from mercury poisoning.

The project on the reduction of trace metals pollution in Meycauayan-Marilao-Obando (MMO) river funded by the Asian Development Bank in 2008- 2009 proposed and actually did pilot projects to reduce metals emissions in smokestacks, did training on simple technologies on pollution control for operators of businesses in the area and conducted educational activities to various stakeholders on the health effects of metals pollution. The Environmental Management Bureau which took over the project from ADB, unfortunately, has not published follow up reports on the outcome of the project, such that no information are available on the present pollution situation of the MMO river system. The bangus, tilapia and various types of shellfishes grown in 15,059 hectares of brackish fishponds in Obando, Bulacan, Paombong and Hagonoy and distributed to local markets maybe contaminated with mercury and lead that can affect consumers of these products. It is imperative for the EMB as part of sound environmental management and for the protection of the health of the general public to provide up to date information on the trace metals pollution of the river environment as well as the contamination data of aquatic products derived from the fishponds which may be affected by the pollution.

Cadmium and soluble forms of arsenic are easily taken up by plants. Knowing that the Philippines had volcanic eruptions that could significantly contribute cadmium and arsenic in farmlands, it would be a good practice for our food regulatory authority to monitor cadmium and arsenic in rice grown from areas which had volcanic eruptions. It would also a good precautionary measure for the food authority to monitor rice that we import since the cadmium and arsenic pollution in the farms where the imported rice were grown is generally unknown. Since rice is the staple food of Filipinos, the regular daily intake of rice could potentially accumulate in the body cadmium and arsenic from rice to levels that can cause toxic effects.

POPs are very powerful toxicants in the sense that they can cause dysfunction of many enzymes and hormones at very small concentrations in the body. In addition, POPs are semi volatile, can stay in vapor form and can be transported to long distances. In effect, the presence of POPs in the environment puts a risk to everyone because they could be present in air which everyone is exposed to. For this reason, it is necessary to eliminate POPs in the global environment and to monitor the residues of POPs in the local environment. Monitoring of POPs in water, soil, biota and air in the Philippines showed that the pesticide residues of DDTs, chlordanes and endrins are still present in our environment despite their banning since the 1980s. Especially disturbing is the presence of high concentrations of chlordane in air detected in residential villages. PCBs and PBDEs are present in our water and sediments which could also be partitioned in air. Monitoring of PCBs and PBDEs in air must be done since the safe disposal of PCB transformer oils and tons of electronic wastes has not been efficiently organized and implemented by the Environmental Management Bureau. Our most pressing environmental and health concern among the POPs are the Dioxins and Furans, the most toxic of the POPs, which are expected to be present in air much more than the other POPs due to the practice of many Filipinos to burn domestic waste and farm waste. Efforts from the government and private sectors to dispose safely materials containing POPs and to stop practices that release Dioxins and Furans have to be improved to prevent further contamination of the environment with POPs.

The most disastrous effect of chemical pollutants to man is the degradation of the quality of the human race. Some mechanisms on how the adverse health effects of chemical pollutants are carried down to the children of exposed persons through genetic and epigenetic effects have been proposed.

The metals cadmium, arsenic, chromium can react with electron - rich bases of the DNA to cause damage to the structure of the gene. PAHs such as benzo(a)pyrene, benzo(a) anthracene,7,12 dimethylbenzanthracene are transformed inside the body by enzymes to strong electron seeking metabolites that can react with electron rich bases of the DNA and cause gene damage. An example of a genetic damage is the mutated breast tumor suppressor gene BRCA-1 that could make the offspring who inherited the mutated gene vulnerable to develop cancer of the breast.

Chemical pollutants can block the expression of a gene without causing damage to the gene through epigenetic effects. Epigenetic effects cause damage to chromatin proteins that are involved in the transcription process in gene expression. Most epigenetic effects are carried through ligand binding of the chemical pollutant to some responsive element in the chromosome which could block transcription. Some chemical pollutants prevent the normal process of deacetylation of histones that signals the opening of the DNA for transcription; or enhance the methylation of DNA binding proteins that results to condensation of the DNA proteins. The aromatic hydrocarbon pollutants PAHs and POPs are implicated in epigenetic effects by binding to the AHR and by activating the AHR-ARNT receptor site in the chromosome (Papoutsis A et al.2010). When the epigenetic effect happens during the critical period of development of the fetus, the defect may be inherited and the effect of the dysfunction of the gene may be manifested in the offspring. An example of inherited epigenetic effect is the blocking of the expression of the breast tumor suppression gene BRCA-1(Papoutsis A et al.2010).

To prevent the increase of NCDs in the country, the mitigation of the exposure of the largest section of the population to known chemical pollutants with NCD effects must be prioritized. A case in point is the exposure of more than 12 millions commuters and residents in Metro Manila to black carbon from traffic pollution which have been shown to have inflammatory (Baja et al, 2013) and cardiovascular effects (Baja et al, 2010). Black carbon is inhalable air particulate in the 2.5 micron size produced by incomplete combustion of fossil fuels. Black carbon would most likely contain toxic trace metals and PAHs which can cause adverse health effects like cardiovascular toxicity, immune suppression and cancer. Environmental and health managers of the country should immediately implement actions to reduce motor vehicle emissions in Metro Manila to prevent the development of more NCDs in the country.

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