Coastal Pollution and Food Resources as Determinants of Population Health in American Samoa

by

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ABSTRACT

Heavy metals and persistent organic pollutants contribute to human health risks worldwide. Among the most common routes of exposure to pollutants for humans are through the consumption of contaminated water and food, with fish being among the greatest vectors for ingestion of heavy metals in humans, particularly mercury.

This dissertation consists of three chapters with a central theme of investigating heavy metal and persistent organic pollutant concentrations in fish and corned beef, which are two commonly consumed food items in American Samoa. A literature review illustrated that historically the primary pollutants of concern in fish muscle tissue from American Samoa have been mercury, arsenic, and polycyclic aromatic hydrocarbon mixtures. To better understand the changes in heavy metals and persistent organic pollutants in fish, this study reports an updated data set, comparing concentrations in pollutants as they have changed over time. To further investigate pollutants in fish tissue, 77 locally caught and commonly consumed fish were analyzed for heavy metals and persistent organic pollutants, and baseline human health risk assessments were calculated for contaminants that had available oral reference doses. While in American Samoa collecting fish for contaminant analyses, it was realized that canned corned beef appeared to be more commonly consumed than fresh fish. An IRB approved consumption survey revealed that 89% of American Samoan adults regularly consume fish, which is the same percentage of people that reported eating canned corned beef, indicating a dramatic increase in this food item to their diet since its introduction in the 20th century.

Results of this study indicate that fish muscle tissue generally has higher heavy metal concentrations than canned corned beef, and that mercury continues to be a main contaminant of concern when consuming fresh and canned fish in American Samoa. While none of the heavy metal concentrations in corned beef exceeded calculated action levels, these foods might contribute to negative health outcomes in other ways. One of the main findings of this study is that either the presence or the ability to detect persistent organic pollutant concentrations are increasing in fish tissue and should be periodically monitored to adequately reflect current conditions.

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To the local farmers and makers for keeping me well fed.

TABLE OF CONTENTS

CHAPTER Page

CHAPTER Page

APPENDIX

LIST OF TABLES

Table Page

LIST OF FIGURES

SAMOAN LANGUAGE CONSIDERATIONS

The Samoan Language has 14 letters (compared to English, which has 26 letters). The consonants are F, G, L, M, N, P, S, T, V. The vowels are A, E, I, O, U.

The letter G sounds like the ‘ng’ in the word song.

All words must end in a vowel.

Many words have glottal stops represented by an inverted comma (koma liliu)

LIST OF ABBREVIATIONS

HMs	Heavy Metals
POPs	Persistent Organic Pollutants
PCBs	polychlorinated biphenyls
PAHs	polycyclic aromatic hydrocarbons (PAHs)
LVPA	Large Vessel Prohibited Area
US-EPA	United States Environmental Protection Agency
AS-EPA	American Samoa Environmental Protection Agency
FDA	Food and Drug Administration
DMWR	Department of Marine and Wildlife Resources
ASPA	American Samoa Power Authority
NOAA	National Oceanic and Atmospheric Administration
CRAG	American Samoa Coral Reef Advisery Group
mg	milligram
kg	kilogram
ppm	parts per million

Dissertation Framework and Hypotheses
Chapter 1

Pollutants, Fishing and Food: Changes in American Samoa

This dissertation begins with an orientation to the study site, briefly reviewing historic fishing data and local pollution data, both of which are related tangentially to contaminants in fish tissue that people consume.

Later, the historical government literature and peer reviewed publications that have measured heavy metal and persistent organic pollutant concentrations in commonly consumed fish in American Samoa were evaluated, then add our current data set for an updated comparison of those contaminants in fish tissue over time, and estimations of associated risk. There are currently five available datasets, plus the data collected for this dissertation. that have evaluated heavy metal and organic pollutant concentrations in local fish tissue. Of those five studies, three are studies initiated by the government and two are from peer reviewed published literature. Mercury is an ongoing contaminant of concern, and overtime there appears to be more measurable concentrations of organic contaminants in fish muscle tissue.

The main objective of this chapter was to summarize the history of pollution research in American Samoa, and to synthesize the currently available heavy metal and persistent organic pollutant data in commonly consumed local fish.

Hypothesis:

I hypothesize that heavy metals and persistent organic pollutants in locally caught fish muscle tissue will currently be at higher concentrations than have been observed from previous studies. Contrarily, if local pollutant inputs have decreased, potentially that will be reflected in the local fish tissue.

Chapter 2

Human Health Risks in American Samoa from the Consumption of Locally Caught Fish Containing Heavy Metals and Persistent Organic Pollutants

For this study, heavy metal and persistent organic pollutant concentrations were measured in commonly consumed fish in American Samoa and created baseline human health risk assessments for each pollutant with an established oral reference doses to assess potential toxicity related to consuming fish that harbor these pollutants. 77 fresh locally caught and consumed fish from the Island of Tutuila were collected and categorized them into four groups based on trophic level dietary preferences. Variations in heavy metal and organic pollutant concentrations were compared between the groups, further identifying if the results of the study are consistent with the published data. Ongoing mercury research should be maintained as it continues to be a contaminant of potential concern.

This data set contributes an update to the limited data on heavy metals and persistent organic pollutants in commonly consumed fish in American Samoa and creates baseline human health risk assessments for all contaminants with oral reference doses.

Hypothesis:

I hypothesize that the levels of heavy metals with oral reference doses in locally caught fish in American Samoa are at high enough concentrations to exceed calculated action levels, particularly mercury in the pelagic species.

Chapter 3

From Wild Caught to Canned Animal Protein: Human Health Risks from the Consumption of Heavy Metals Found in Canned Meats Frequently Consumed in American Samoa.

While fresh local fish is still a staple in the (American) Samoan diet, in recent years canned tuna and canned corned beef have become regular food sources for most people of American Samoa. When local folks were surveyed, the same number of respondents report regularly consuming locally caught fish as they consume canned corned beef (89%), and canned tuna is common largely due to the presence of the local tuna plant. Here I measured and compared Heavy metal concentrations were measured and compared in canned tuna in oil and in canned corned beef and a baseline human health risk assessment was calculted. Further heavy metal concentrations were compared between canned and fresh tuna. Reporting heavy metal concentrations in commonly consumed food items allows people to make more informed choices regarding the foods they choose to consume.

Hypotheses:

I hypothesize that wet weight tuna will generally have higher heavy metal concentrations than the equivalent mass intake of wet weight corned beef, particularly from mercury, and that fresh tuna will have higher concentrations than the equivalent mass intake of canned tuna, except for potentially biologically necessary elements in found in bovine muscle tissue.

INTRODUCTION

Heavy metals and persistent organic pollutants are present in ecosystems world-wide creating potential health risks to the people who are exposed to them. Island communities experience unique exposure to a range of environmental pollutants, from macrodebris to micropollutants (Halpern et al. 2008). Islands, particularly small ones, have relatively minimal land area for waste disposal and high associated cost of exporting garbage. Land based garbage sources, along with global ocean patterns, result in trash pooling and gathering along coasts (Elliot and Elliot 2013), and generally island communities rely on the ocean for their livelihood and resources.

Exposure to heavy metals and persistent organic pollutants might also be elevated on island due to natural and anthropogenic causes (Rick et al. 2013). Geologically, volcanic islands inherently have elevated concentrations of some metals, such as arsenic and cadmium (AS-EPA 1991). Land use change and other human activity further liberate these metals into the environment. Industrial activities that emit toxic chemicals often occur along waterways, where uncontrolled effluent might enter the water where it will then either absorb into the sediments or drain and disperse into coastal systems. The nature of water flowing downhill dictates that without some form of mitigation, pollutants will end up entering the ocean, and further into fish and other organisms. Heavy metals and persistent organic pollutants can be bound to the sediments or in the biota in the environment, entering the food chain, potentially creating toxicity for the individuals consuming them. Long distance atmospheric transport and deposition also contributes to overall chemical distribution globally; this is particularly noticeable when evaluating global mercury concentrations (in the ecosystem and in fish), which largely results from coal burning for energy purposes.

Island life often dictates that the inhabitants are (or at least were) likely dependent on the ocean for resources, and often are dependent on coastal seafood as a major protein source. Among the primary routes of exposure to heavy metals and organic pollutants in humans is from the food items that they consume, particularly fish (Bosch et al. 2015, Schukla et al. 2022, WHO 2023). Fish tend to contain among the highest concentrations and greatest diversity of heavy metals in their tissue when compared to other animal protein sources, and the trends indicate that many metals biomagnify in higher trophic species, including humans.

This study focuses on American Samoa, a small, group of isolated islands in the South Pacific Ocean. Most of the human population lives on the Island of Tutuila, which is also the island most impacted by human activity and pollution (DMWR 1984, AS-EPA 2006). Traditionally the people of American Samoa have relied heavily on fish as a major food and protein source, but as they have become incorporated into the global economy in the past 100 to 150 years, fishing effort and fish consumption has decreased (Craig et al. 1999). Progressing along a similar timeline, the people of American Samoa have been exposed to food subsidization from the United States, New Zealand, and Australia, creating an environment with novel high fat low nutrient dense foods which have contributed to shifts in dietary preferences and overall health outcomes of the community (BBC 2016; Errington and Gewertz 2008, Neuendorf et al. 2021). This study investigates Heavy Metal and Persistent Organic Pollutant concentrations in commonly consumed fish and popular canned meats in American Samoa to assess baseline human health risks based on these metrics.

Over the course of two years, 2017 and 2018, I traveled to American Samoa for three trips, approximately two months each trip. I immediately started interacting with the local government and local agencies to figure out how best to conduct research in a way that respects the community and local authority. I entered communities and got to know local people, asking for their permission and blessings to collect sediment, water, and biota samples. The people were so kind to me; I was invited to celebrations and to weekend BBQs. Interactions with these lovely people illustrated the many challenges that the people of American Samoan encounter and allowed for me to contextualize the importance of considering food choices. I started this project to assess risks associated with consuming heavy metal and persistent organic pollutants in fish, but realized the greater risk might be from novel canned food items, not from fish at all.

CHAPTER 1

POLLUTANTS, FISHING, AND FOOD: CHANGES IN AMERICAN (AMERIKA) SAMOA

Abstract

Humans are impacted by environmental degradation world-wide. Factors such as habitat destruction, pollution, and excessive resource exploitation reduce ecosystem and human health; with isolated island communities being particularly susceptible to local impacts and interactions (Elliot and Elliot 2013, Halpern et al. 2008, Rick et al. 2013, Vikas and Dwarakish 2015). Life in American Samoa, a small island in the South Pacific Ocean, has changed dramatically in recent history, moving from living largely in a state of isolation in the early to mid 1800s to being the site of an active U.S. Naval Base and a Territory of the United States by the turn of the 20th century. As American Samoa became more incorporated in the global economy, they were also exposed to new technologies and foreign goods. This study discusses the historical context of fishing practices and chemical pollutants in American Samoa and presents a review of heavy metals and persistent organic pollutants measured in commonly consumed locally caught fish.

In response to the increasing effects of environmental pollutants the United States established the Environmental Protection Agency (US-EPA) in 1970. The first water quality studies were conducted in 1979 and investigated nutrients such as nitrogen and phosphorus. In 1990, the American Samoan Government conducted the first documented evaluation of heavy metals and persistent organic pollutants in fish that were commonly

consumed by local people. They identified potentially elevated levels of arsenic (As), lead (Pb), mercury (Hg), the polychlorinated biphenyl (PCB) mixture Arochlor 1260, and DDD and DDE (in one individual fish). These results guided future studies in American Samoa. After teasing out natural (vs anthropogenic) sources and identifying the toxicological differences between various forms of the contaminants of concern, by 2015 the only contaminant being measured in local fish was mercury (Hg).

The heavy metal results from the current study are consistent with findings from previous research, identifying mercury (Hg) as the most pressing metal of concern when consuming local fish. This study identified more persistent organic pollutants in fish tissue than previous studies have, including measurable concentrations of multiple polychlorinated biphenyls (PCBs), polycyclic aromatic hydrocarbons (PAHs), pesticides, and phthalates.

Introduction to the study site, American Samoa

American Samoa is a group of five small remote islands and two atolls in the South Pacific (14.2710° S, 170.1322° W), roughly between Australia and Hawaii, and about 1,000 miles south of the equator. Tutuila (Figure 1.1), the largest island of American Samoa, has a total area of approximately 142 km^2. There are approximately 49,710 human inhabitants based on the United States Census Bureau (2020), which comprises approximately 95% of the territory's population, and is the hub for commerce and industry for all of the islands. The closest large land masses to American Samoa are New Zealand and Australia, at a distance of 3,295 km and 5,951 km respectively.

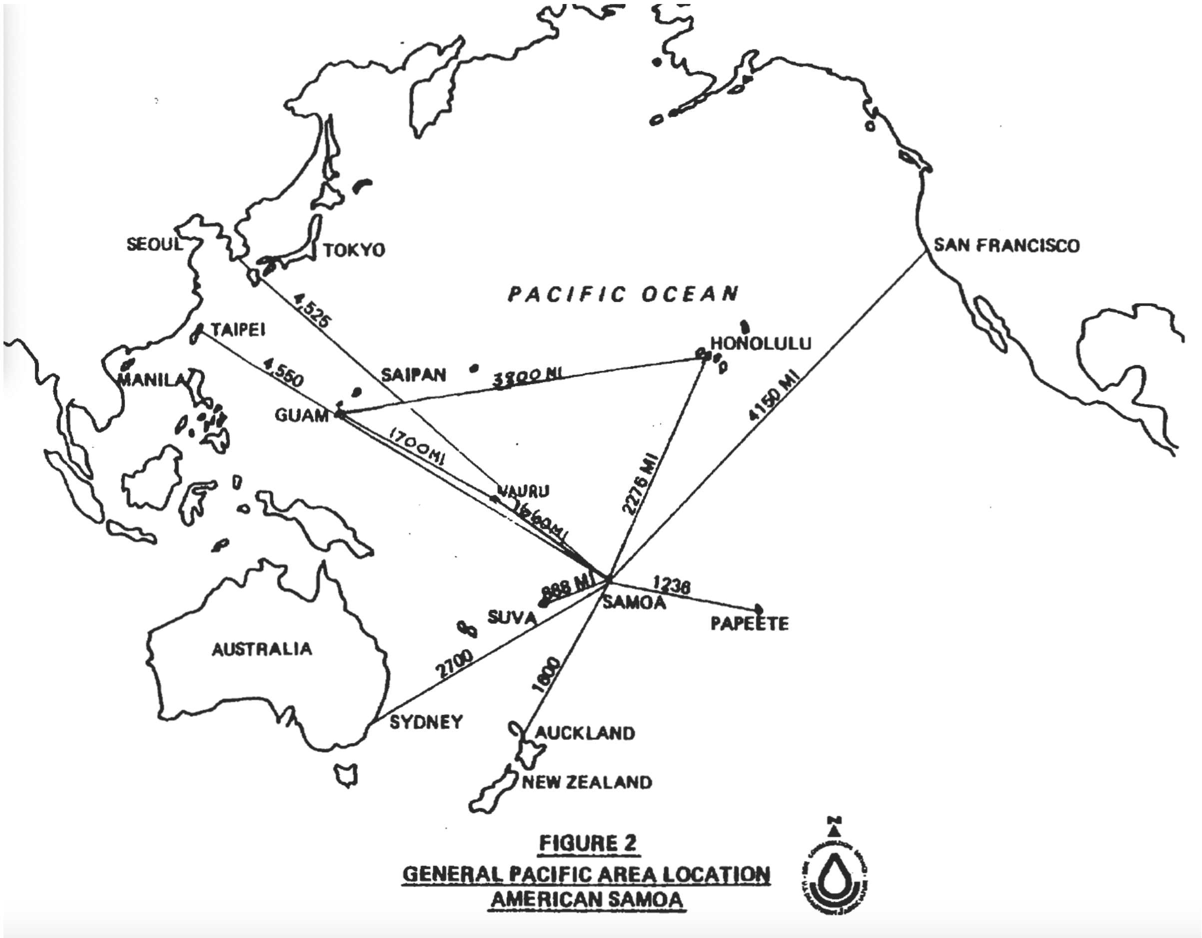

Figure 1.1
Location of (American) Samoa in the South Pacific Ocean (From: Perry (1986))

Archaeologists estimate that people first arrived at the Samoan islands ~3,000 years ago, based on unearthed pieces of pottery, and stone tools and remnants of food items such as reef fish, clams, and snails have been dated up to ~1,500 years ago (ASHPO 2020). European explorers began documenting interactions with native people of the Samoans islands in the early to mid 1700s, but contact remained minimal until the 1830s when missionaries and traders began frequenting the Samoan Islands, bringing along with them new ideals, technologies, and desirable goods (US Embassy in Samoa 2023).

Pago Pago Harbor on the island of Tutuila is one of largest and deepest harbors in the world, providing shelter from the wind and seas from the open ocean, making it a highly desirable staging location for sea travelers (Britannica 2023). As maritime expansion and commerce expanded in the mid to late 1800's into the Pacific Ocean, major world powers were staking claims on Pacific Islands. In response to the encroachment by Germany, Britain, and the United States, in 1872 the local Matai (chiefs/ leaders) reached a treaty with the United States government where in exchange for protection from other potentially aggressive adversaries and access to more resources, the U.S. Navy was allowed to develop a coaling station and Naval Base in Pago Pago Harbor (National Park Service 2023). In 1900 American Samoa officially became an unincorporated United States territory (United States Department of the Interior 2023). The U.S. Navy built infrastructure, introduced novel goods, and was an active presence in American Samoa until 1951 when the Naval Base was shut down and active military operations largely ceased (Campling and Havice 2007). Authority of the territory was transferred to the Department of the Interior (from the Navy) in 1956, and in 1967 local Samoan authorities created their own constitution, highlighting a government mostly controlled locally, but with general mainland U.S. laws and guidelines (U.S. Department of the Interior 2023).

Brief History of American Samoa as a Fishing Community

American Samoa is historically a fishing community, relying on local seafood resources as a rich protein source. Being completely surrounded by deep ocean waters, traditional foraging was reliant on small wooden boats that were required to remain close to shore. In a location where food resources are limited, people historically relied on the ocean as a major source of food, particularly species foraged or caught very near shore. Overtime fishing technology became more complex, and local people were able to purchase and utilize larger fishing vessels that could travel further offshore.

Traditional subsistence fishing practices were primarily conducted from shore or occasionally from small wooden canoes, known as va’aalo or bonito canoes (Indigenous Boats 2013), and fishing was a family affair. Local villages maintain control of the fishing resources adjacent to their land (Levine and Allen 2009), and generally discourage trespassing without distinct permission. Precolonial fishing activity was closely tied to cultural customs and seasonality, and was conducted to benefit the community, with effort particularly during times of celebration (fa’alavelave). Fish catches were shared and distributed according to the village hierarchy, first to the matai (chiefs), then to the faife’au (ministers), then finally to one’s extended family and neighbors (Kleiber and Leong 2018). Fishing and sharing one’s catch is traditionally regarded as an important part of Fa’a Samoa (The Samoan Way).

As American Samoa became more incorporated into the global economy, fishing practices started to move away from coastal community collection methods to broader open ocean methods. Foreign fishing vessels started bringing commercial catches of tuna to AS in the 1950s, when the first tuna cannery opened in Pago Pago Harbor (Kleiber and Leong 2018). Until the 1970s, the most prominent fishing vessels were paddle operated paopaos (small outrigger canoes) (Figure 1.2) and were limited to near shore activities. In the early 1970s, local fishing operations were funded for modernization, upgrading to a fleet consisting of plywood catamarans and welded aluminum vessels (alia) which were constructed to allow for greater exploitation of offshore marine habitats and access to more commercial fishing practices. Over time the wooden vessels were largely replaced with larger motorized aluminum vessels, limiting traditional vessels to times of calm weather and celebrations.

Figure 1.2
Samoan Paopao – Traditional Fishing Vessel. (Image: by Tiffany Lewis 2018)

The new fleets engaged heavily in bottom fishing, until diminishing catches and changes in skilled fishing practices directed efforts to focus on trolling specifically for pelagic species (WPRFMC 2019). As fishing practices increasingly shifted to local longline commercial fishing, AS was further incorporated into the market economy. The late 1990s saw a rapid increase in longline fishing. Monohull longline vessels, capable of catching tuna and other large pelagic fish began to dominate fishing practices. To reduce competition between small and large vessels, a Large Vessel Prohibited Area (LVPA) was established in 2002 and dictated that longline fishing boats >50 ft. in length were restricted from fishing in certain areas to reduce competition with smaller operations and to support decreasing fish populations (Kleiber and Leong 2018).

Fishing practices were further complicated in 2009 when a devastating tsunami damaged or destroyed boat docks and over 50% of local small fishing vessels, resulting in an estimated decrease of 80% in bottom fishing revenue (Wallace 2019). When the LVPA was developed, there were approximately 40 (25-40ft.) alia longline catamaran vessels in operation. In 2014 only one alia longline vessel was reported to be in operation, which has led to an allowance of large vessels to operate within the LVPA to support the AS fishing economy (WPRFMC 2016). At its peak, the number of large longline vessels totaled 66 in 2001/2002, but by 2018 the total was reduced to 13. This time has also seen an overall reduction in catch per unit effort (CPUE) (FRMD 2019). Although catch estimates have been calculated for commercial landings, the demography and landing data of reef and bottom fish species is poorly understood, therefore translating landing data to fish abundance data is difficult. Currently, local tuna fishermen sell their catch to the local Starkist cannery, to local restaurants and markets, and leftovers to locals at roadside stands. Pelagic fishes including tunas are require larger boats and more fishing gear. They are sold to the local Starkist cannery, to local on island restaurants, and along the one main road on Tutuila. Fresh caught tunas are cost prohibitive for many families.

Long before sea faring vessels were introduced to American Samoa, most seafood consisted of species found close to shore. Bottom and reef fishes, such as Lined

Surgeonfish (*Acanthurus lineatus;* Alogo (Samoan Name)), Big-scale Soldierfish (*Myripristis berndti;* Malau-ugatele/ Malua-va'ava'a), and Yellowstrip goatfish (*Molloidichthys flavolineatus;* Afolu) are commonly caught using shore based or small boat operations (AS Creel Survey 2018). They are taken home for personal consumption, as well as sold to local stores and at roadside stands along the single main road (Route 1) in American Samoa. Reef and bottom fishes are caught both by commercial vessels and using traditional practices and are primarily eaten at home and sold at local markets and at roadside stands. Economic change, access to imports, and diminishing local seafood resources (Craig 2004) have led to a change in effort for traditional fishing practices, are resulting in evidence of overall reduced fishing effort over time (Craig et al. 1993, Levine and Allen 2009; NOAA 2018)

The two most impacted and researched sites in American Samoa are Nu'uuli (Pala) Lagoon and Pago Pago Harbor. 44% of the population of Tutuila live in villages that border these two sites, and the bulk of industry and commerce occurs here (Richmond 1995).

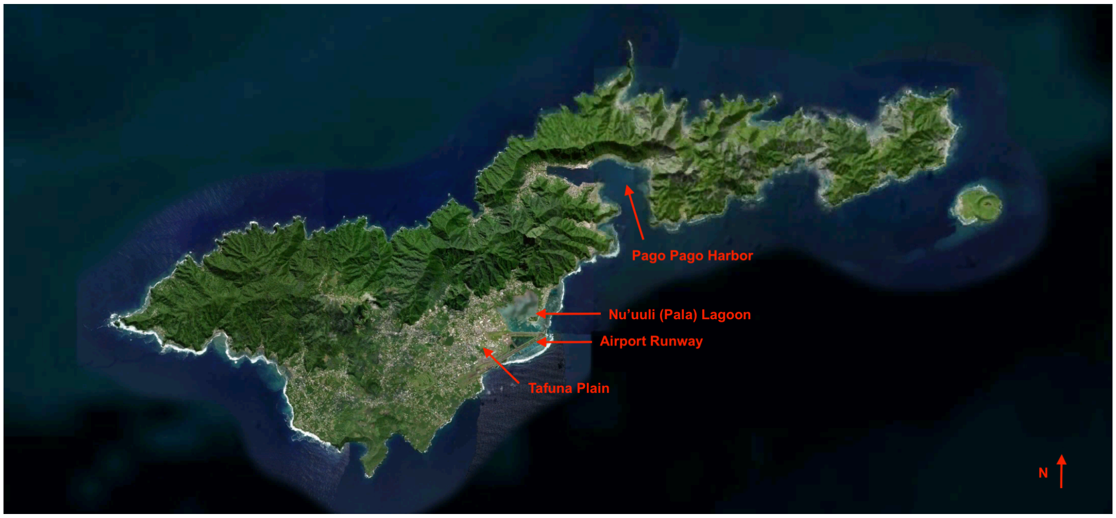

Figure 1.3
Pago Pago Harbor and Nu'uuli Lagoon: Hubs of Pollution Impact and Research on Tutuila
Island of Tutuila, American Samoa (From Google Maps 2023).
Highlighting Pago Pago Harbor, Nu'uuli (Pala) Lagoon (with the airport runway), and the Tafuna Plain.

Pago Pago Harbor

Pago Pago Harbor (Figure 1.2) is ultimately the reason for the establishment of American Samoa as a United States territory. The harbor is large at approximately 4km long and 1km wide, and the water is deep ranging from 120 to 200 feet (CH2MHILL 1984) allowing for large ships to gain shelter from potentially dangerous sea conditions, in a location surrounded by vast ocean expanses. These characteristics made it ideal as staging and refueling area for ships. At the turn of the 1870's local chiefs allowed the U.S. to set up a coaling station to fuel U.S. ships as they traveled through the Pacific Ocean (Anderson 1978) and in 1872 the U.S. was allowed exclusive rights to Pago Pago Harbor in exchange for protection from invasion by Britain and Germany, and greater access to resources. American Samoa officially became a US territory in 1900. At this time, the US Navy built a naval base with several modern buildings, and Pago Pago

Harbor was used by the US Navy as a coaling and repair station. It was utilized as a base in the South Pacific until Navy operations mostly ceased in 1951 when the Navy Base officially shut down (Campling and Havice 2007). Military operations have contributed to ongoing environmental concerns because military waste has been dumped at unknown sites around the island (Thomson and Samuels-Jones 2022). Although military operations have ceased, there are toxic materials of unknown location throughout the watershed and around the island that leak chemicals and present mechanical hazards. It is rumored that there is military waste buried under an active elementary school. The military waste includes munitions and toxic chemicals in including plutonium, dioxins, nerve agents, and live munitions, but historical records of types and location of buried materials were not created or maintained; therefore, clean-up is nearly impossible at this (Thomson and Samuels-Jones 2022).

The harbor has approximately 27 streams that drain into it, with a high potential for run-off due to relatively low substrate permeability, and steep slopes reaching grades up to 60% (Davis 1975, US-EPA 2006). This runoff transports garbage and other pollutants downstream to concentrate along the shore (Figure 1.4). Homes and businesses are relatively densely packed along the shores and upslope from the harbor (Richmond 1995). Some of the homes have small agricultural operations and pig pens, which also result in potentially toxic pollutants being distributed into the harbor from run-off. The harbor is the major point of commerce, industry, and government for all the American Samoan islands (US-EPA 2006), with the most prominent industry currently being the tuna canneries. Because of the large amount of boat traffic, there is a shipyard within the harbor that services local and foreign boats, including paint maintenance and repair (US-

EPA 2006). Over the years many boats have sunk in the harbor, many loaded with fuel and other chemicals, many of which still remain sunk and leaching (Craig 2004)

Figure 1.4
Large metal debris in a stream entering Pago Pago Harbor. Garbage along the shore in Pago Pago Harbor (Images take by Tiffany Lewis 2017)

Most studies regarding pollutants in American Samoa have been conducted in Pago Pago Harbor due its high levels of human activity and impact. Approximately 95% of the harbor habitat has been irreparably altered to accommodate human activity. 100% of the mangrove stands have been converted for human use, which has reduced the ecosystem's natural ability to retain sediments and filter contaminants and has eliminated nursery habitat for fish and other organisms (Craig 2004). Among the first pollution related scientific assessments in the 1980's in Pago Harbor concluded that most of the pollution at that time was attributed mostly to contaminant mobilization from runoff, effluent from the tuna canneries, and ongoing sewage outfall (CH2MHILL 1984). Craig

(2004) reported that due to the chronic inputs of toxic chemicals, the harbor is considered an ‘environmental write-off’.

Tuna Canneries

The tuna cannery business is the greatest source of revenue for American Samoa and employs the greatest number of people there (US Government Accountability Office 2023). In 1954, the United States fish processing company Van Camp (also known on island as Samoan Packing, and internationally as Chicken of the Sea) built the first tuna processing plant in Pago Pago Harbor. This was in response to the expanding tuna fisheries; there were masses amount of tuna that needed to be processed for human and pet food, and they needed infrastructure to process it all. With the reduction of jobs from military operations (reminder: the Navy base closed in 1951 and the first cannery opened in 1951), the cannery filled an economic void. Business was booming for the tuna industry, which prompted a second tuna processing plant and cannery to open on island. In 1963, the Starkist Tuna plant began operations in Pago Pago Harbor, adjacent to the Van Camp plant (Campling and Havice 2007, Levine and Allen 2003). Both canneries operated until the Van Camp (Chicken of the Sea) plant shut down in 2009/ 2010 due infrastructure damage, and legal and financial trouble. The tuna cannery operations are still in decline due to diminishing demand and financial challenges (The U.S. Government Accountability Office 2023)

Nu'uuli Lagoon

Nu'uuIi (Pala) Lagoon is a ~50 ha coastal mangrove swamp located in Tutuila, American Samoa. It is the largest remaining and most threatened wetland and mangrove habitat on Tutuila (Volk 1993) (Figure 1.3). The lagoon is approximately circular in shape, with a diameter of approximately 1.6km^2 to 2 km^2 and depths ranging from 1-5 meters depending on the tides. Fresh water streams run into brackish mudflats converging with saltwater at the mouth of the lagoon. The spatial variability of water characteristics and geological structure result in multiple habitat types, ranging from coral reefs at the ocean's edge to mangrove lined mudflats and rocky streams. Nu'uuli Lagoon houses the largest remaining stand of mangrove forest in American Samoa, including the largest stand of red mangrove, *Rhizophora mangle* and the potentially threatened puzzlenut tree, *Xylocarpus moluccensis*. As a mangrove lagoon habitat, Nu'uuli lagoon provides nesting and feeding grounds for coastal birds, nursery habitat for many land and marine species, and ecosystem services to humans including food and recreational opportunities (Yamasaki 1985).

As of 1993, 33% of Nu'uuli's wetlands had been directly converted for human use (Volk 1993). And one of the major land use changes in the lagoon was the construction of Pago Pago Airport in 1956, which was constructed over coral reef habitat, and reduced the outflow area of the lagoon by approximately 66%, which affected water exchange and flushing times. Reduced flushing results in higher instances of water stagnation and the potential for pollution retention (Volk 1993).

Although people picnic and recreate in between the airport runway and one of the major streams feeding the lagoon (Papa Stream at Lions Park), they generally stay out of the water based on its long-standing reputation for being heavily polluted (personal observation, personal communication with multiple locals in AS, Yamasaki 1985) (Image 1.3). Since 1985, the abundance of single use plastics, Styrofoam food containers, and overall increase of disposable products has dramatically added to the pollution problems (NOAA 2017), personal observation) to a system with few resources to get rid of it. Nu'uuli Lagoon was designated as a Special Management Area by the Coastal Management Act of 1990, but lagoon health has continued to decline.

The lagoon health is tightly coupled to human behavior and well-being, and the lagoon is less able to provide the services as it has in recent past due to environmental degradation and pollution. It is recognized that successful management strategies in Nu'uuli Lagoon will require participation by the community and relevant village councils (Volk 1993). For decades community members have expressed their concern about the increasing levels of toxic pollutants in Nu'uuli Lagoon, and its overall health (Yamasaki et al. 1985, personal communication with residents of the lagoon and local government and nongovernment agencies), but they do not yet have tangible solutions to make lasting positive impacts towards less debris and overall pollution.

Figure 1.5
Macrodebris in Papa Stream at Lions Park in Nu'uuli Lagoon (Images by Tiffany Lewis 2018)

The Illusive Clams of Nu'uuli (Pala) Lagoon

Nu'uuli (Pala) Lagoon supports diverse populations of invertebrates, fish, and birds, as well as the local people (Volk 1993, Yamasaki et al. 1985). Historically the lagoon was identified as a 'major source' of income and food for Nu'uuli and Tafuna village residents because of its various species of fish and invertebrates, including two species of smal edible clams (*Gafrarium tumidum* and *Asaphis deflorate;* Tunage and Pipi*)*. Ponwith (1992) reported that "clamming was a popular fishing method"; clammers would search for syphon holes visually at low tide or probe with their bare feet, then dig holes with a knife to extract them. *G. tumidum* was reportedly abundant along the north and eastern boundaries of the lagoon (Ponwith 1992, Yamasaki et al. 1985). *A. deflorata* was collected in sandy substrate along the airport runway and at the South end of Lions Park by probing with an available tool to expose individuals buried in sand ~5-30cm deep (Ponwith 1992). In 1985, 91% of families surveyed reported that they searched for clams an average of 127 days per year and 64% searched for crabs 109 days per year (Yamasaki et al. 1985). In 1991, Ponwith (1992) estimated the total yearly clam catch (of *G. tumidum* and *A. deflorata)* to be 5,448 lbs from Nu'uuli Lagoon; the majority being of *G. tumidum* for a total of 4,638 lbs. They report that 100% of *G. tumidum* were collected from residents of Tafuna or Nu'uuli; *G. tumidum's* reported distribution corresponds to private property. 93% of *A. deflorata's* catch was reportedly collected by Tafuna or Nu'uuli residents, with a reported distribution along public and private properties. 62% of *G. tumidum* and 78% of *A. deflorata* were kept for family consumption, and the rest were sold to local consumers (Ponwith 1992).

The current populations of *G. tumidum* and *A. deflorata* are unclear, even to the people who are most affected by its fluctuations; the locals who used to clam, and those who consumed them. When I began investigating the clam populations in Nu'uuli Lagoon in September 2017, I was unable to find a single clam, nor was I able to acquire even one harvested clam nor any current active clammers. I only encountered one person hunting in the lagoon, but for crabs, with no success. Ultimately, the status of the clam populations and the future of the clam fishery is unclear and appears to be currently stagnant. When I asked people about the clams, the most common response was to tell me that they should be there, and that I could find people selling them along the road. They were surprised then to hear that they were hard/ impossible to find at this time. In 2020, a professor from the American Samoa Community College contacted me letting me know they harvested some clams to eat and that they plan to eventually put together an American Samoa guide to bivalves, otherwise I have been unable to find any updated information regarding the *G. tumidum* and *A. deflorata* population or the fishery.

Pollution in Nu’uuli Lagoon

Locally, Nu’uuli Lagoon is also known as Pala Lagoon. In Samoan, the word ‘Pala’ literally translates to dirt, rotten, and decay in Samoan. These terms are indicative of how the wetlands are often perceived as wastelands in American Samoa. Anthropogenic macro-debris is common and diverse throughout the lagoon. Trash was often used as a base fill when building homes, identifying a particular area behind a store named South Pacific Traders which has been cleared and filled using trash and cinder blocks (Yamasaki 1985). Areas of the lagoon have been allocated as unofficial landfills (Volk et al. 1993, Yamasaki et al. 1985), where items from common household waste to appliances to vehicles are deposited. Fabrics and wires are woven through mangrove roots (Image 1.6). Metal fragments, building materials, and appliances litter the ground in varying forms of decay, and are buried deep in the mud, creating mechanical and chemical hazards. It is also common to see overflowing dumpsters sitting along water’s edge (Image 1.7). I have personally witnessed people disposing of waste directly into the creeks on multiple occasions, which is a historic behavior from when food items and waste were biological and more biodegradable. To facilitate waste management the American Samoa Power Authority (ASPA) offers solid waste pick up for a fee, which is then transported to the Futiga landfill (Van der Ryn et al. 2016).

Figure 1.6
Fabrics and garbage stuck on mangrove roots. (Images taken by Tiffany Lewis 2018)

Image 1.7
Overflowing Garbage Cans Along the Coast on Tutuila (Image by Tiffany Lewis 2017)

In addition to direct deposition, run-off from rainfall collects and transports debris and chemical pollutants down-stream, flushing into two main streams flowing into Nu'uuli Lagoon, Papa and Vaitele. Styrofoam food containers, plastic bottles, and flip-flops are commonly identified island-wide, concentrating at mouth streams and other areas of convergence (NOAA 2017, Ve'e and Comeros 2016). Chemical pollutants have also been identified in the water and sediments in this system (Polidoro et al. 2017). In 2015, samples from Papa and Vaitele streams were found to contain heavy metals and several categories of organic contaminants, including pesticides, PAHs, and phthalates

(Polidoro et al. 2017). Among the most toxic of the chemicals identified is parathion, a pesticide which was banned by the US Environmental Protection Agency in 2003.

Garbage and Pollutants on Tutuila

Sources of Garbage on Island

As foreign occupation, land use change, and access to imported goods dramatically increased in the 20th century in American Samoa, environmental degradation and the effects of pollution became increasingly more evident.

Pago Pago Harbor regularly receives large container ships of material goods, leaving behind waste which rarely leaves the island. With limited land area, waste disposal in American Samoa has many challenges. There is one unlined landfill on Tutuila in Futiga, which was expected to reach capacity in 2015 (Van der Ryn et al. 2016), but due to lack of space and limited options, the American Samoa Power Authority (ASPA) suggested that the garbage be recompacted to extend the life of the landfill until further notice (BBC 2016). There is one recycling staging facility that accepts clean aluminum cans (which are shipped off island) and for Vailima (local Samoan beer brand) beer bottles, which can be cleaned and submitted for reuse in Independent Samoa. Garbage collection services are cost prohibitive for many families, and illegal dumping and littering are common, resulting in trash aggregations in communities and along coastal areas. Burning is a common method for waste disposal in American Samoa, which can create toxic fumes depending on the incinerated material type and local conditions, but due to winds doesn’t tend to remain in the air. It is also common to see items such as cars and household appliances haphazardly disposed of on land and in near-shore marine areas (visual confirmation; Yamasaki et al. 1985), which can leach chemicals into the environment over time. Consequently, there are potential chemical hazards from improper waste disposal.

For early Samoan communities, it was historically common to dispose of unwanted or used items, such as coconut husks and fish waste into streams to be ‘taken away’ (Brown 2015). These practices still readily occur, but due to the rapid introduction of non-biodegradable materials and invisible chemicals, coastal ecosystems are negatively impacted. The impacts of macro debris are visible and thus more readily identifiable as being potentially problematic. It is common to see large debris when swimming in the ocean or walking along the coast. There are many instances around island where macro debris, such as electronic waste, old appliances, and sunken ships leak toxic chemicals into the surrounding environment. One of the most known instances of macro debris leaking chemicals is from the gasoline tanker known as the *USS Chehalis*, which exploded and sunk in Pago Pago Harbor in 1949 near the fuel dock in approximately 160 feet of water. Fuel has been leaking steadily from the ship since then, and there is concern that as the ship deteriorates greater amounts of the up 600,000 gallons of fuel currently on board the ship will be liberated, of which the fuel tanks are largely still intact (AS-EPA 2006).

Nitrogen and Phosphorus: The Early Days of Studying Environmental Pollutants in American Samoa

In 1970 the United States Environmental Protection Agency (US- EPA) was established, and thus most of the official research studying environmental pollutants in the United States have occurred in the past approximately 50 years since the establishment of the US-EPA.

In response to the Federal Water Pollution Control Act amendments of 1972 and the enactment of the Clean Water Act in 1977, the first official comprehensive (for the time) water quality study for Pago Pago Harbor occurred in 1979 by the United States Army Engineer District based in Honolulu, Hawaii on. The goals of the analyses were to evaluate baseline levels of total phosphorus (TP), total nitrogen (TN), and to investigate turbidity for salt and freshwater conditions. TP and TN are essential nutrients, but at elevated levels they can promote excessive algal growth and reduced oxygen conditions once the blooms decay, promoting fish die offs (US – EPA Nutrient Pollution 2023). Chlorophyll-a and compensation depth were analyzed for ocean sites, and suspended solids and fecal coliform were analyzed for streams within the Pago Pago Harbor watershed since it was site most impacted by human modification. Samples were collected from various sites around Pago Pago Harbor, and it was determined that 75% of the TN and TP inputs were from the tuna canneries, and the remaining 25% of TP and TN inputs was from sewage outfall and nonpoint stream sources. Excess chlorophyll-a and low dissolved oxygen were estimated to be at levels unlikely to support diverse biological communities. In response to this study, it was recommended that the cannery discharge be relocated to outside of the harbor where there is greater water movement allowing for greater dilution of TP and TN, and they recommended building chlorination facilities to support any additional discharge activities in the area (AS-EPA 1979). Note that this study estimated that approximately 75% of anthropogenic TN and TP inputs are from the canneries, and approximately 25% being from sewage outfalls (and unknown non-point sources). These results largely guided future research on island, and subsequent action by the local government.

Currently, the American Samoa EPA (AS- EPA) frequently samples and tests beaches around the island and within Pago Pago Harbor to monitor for N and P levels, and 46 recreational beaches are also regularly sampled and tested for bacteria levels (AS-EPA Water Quality Monitoring 2023).

Waste from The Canneries

From 1954 when the first cannery opened until 1990, fish waste from the two operating canneries was disposed of within the harbor. The effluent contained high levels of nutrients (including nitrogen and phosphorus) and oil and grease (Starkist Samoa Company 2021) which can lead to algal blooms and low oxygen conditions. Due to decreasing water quality 1990, the two tuna canneries collaboratively built infrastructure to begin pumping high strength fish waste to an outfall outside of harbor, which was estimated to reduce nutrient output into the harbor up to 60% (CH2M HILL 1991).

In 2009 a devastating tsunami (dubbed the Samoa Tsunami) caused major infrastructure damage to the Chicken of the Sea plant, which prompted the plant to close. In 2012 the Starkist plant informed the EPA that they would not be repairing the infrastructure to pump the waste out of the harbor, and instead would route it through onsite treatment equipment and once again discharge the waste though the original outfall into the harbor, which is still currently occurring (Starkist Samoa Company 2019). As recently as 2019, the EPA fined the Starkist cannery for failure to comply with effluent regulations (US – EPA 2019).

Sewage Outfalls

In response to the measurements of elevated levels of total phosphorus and total nitrogen from the 1979 AS-EPA study, research was initiated to evaluate the physical, chemical, and biological characteristics of sewage outfalls in Pago Pago Harbor, as well as to investigate sites that might be more suitable for discharging wastewater (DMWR 1984). The scientists noted that the sewage outfall plumes were visibly surfacing during sampling events and identified that phytoplankton densities (measured as chl-a) were generally up to 100 times higher in the inner harbor nearer the plumes and had a gradient to lower densities near the outer harbor where there is greater water movement and ocean mixing leads to greater dilution. In response to the 1984 study, improvements were made to the sewage processing plant resulting in piping sewage waste outside of the harbor (ASPA 2019).

Currently there are two sewage treatment plants (Utelei and Fogogogo) which are controlled by the American Samoa Power Authority (ASPA) and overseen by the US – EPA. They are required to perform regular testing of the waters in proximity to the outfalls, and every five years the treatment plants must re-apply for permitting (US – EPA 2019).

Tafuna Plain: Pollutants in Household Water

In 1974 the AS government started to create wells for groundwater storage to support fresh water sources for the growing human population, particularly on the Tafuna plain due to its flat expanses (Figure 1.2). These wells became a main household and drinking water source, but frequently had been found to have elevated turbidity and coliform bacteria resulting largely from animal and human wastes, particularly after times of heavy rainfall (Kennedy et al. 1987). While I couldn't find the exact details of future wells, the USGS reported increasing numbers of wells are reaching spatial capacity and the more wells per area increases the likelihood of contamination (USGS 2007).

Identifying Heavy Metals and Persistent Organic Pollutants in American Samoa

Among the earliest metals of potential concern identified in Pago Pago Harbor were arsenic (As), chromium (Cr), Copper (Cu), nickel (Ni), and zinc (Zn) in 1977 by the EPA (AS-EPA 1991), but similarly "high" levels of these metals were reported in Hawaii where human induced pollution impacts were considered to be minimal, thus it is hypothesized some metals might just occur due to naturally occurring geological weathering; it is possible that background levels of metals might exceed toxicity criteria, but it is challenging to distinguish between background levels and those which are introduced or liberated by human activity. It is further challenging to determine true background levels because environmental sample collection and contaminant measurements started after pollutants had already been introduced into these systems, and irreparable land use change had occurred (Craig 2004).

Heavy Metals and Persistent Organic Pollutants in Fish Muscle Tissue

The Agencies Involved

In response to visible evidence of pollution, including algal blooms and fish die offs, the American Samoan and Federal United States (U.S.) governments began periodically investigating environmental samples for biological and chemical pollutants, and sources of those pollutants around the mid 1900's. In 1970 the U.S. Environmental

Protection Agency (US-EPA) was established which initiated formal research studies to evaluate local pollution levels and the potential hazards associated, and to establish protocols and action plans to mitigate potentially hazardous conditions. The US-EPA has a mainland United States branch and an American Samoan (AS-EPA) branch. Together, they have been the leading agencies to address local pollution issues, along with other agencies, including the National Oceanic and Atmospheric Administration (NOAA), the Department of Marine and Wildlife Resources (DMWR), and the American Samoa Coral Reef Advisory Groups (CRAG).

Methods and Materials

Literature Review

For this review, the commonly available data regarding heavy metal and persistent organic pollutant concentrations in fish were located by performing literature searches through the Arizona State University libraries and Google Scholar. The search was conducted to include all years, and included the search terms: American Samoa, heavy metals, persistent organic pollutants, mercury, arsenic, lead, health risk, fish. Among the most valuable sources of literature were provided to me by local agencies in American Samoa, some of which cannot be accessed using available search engines. Because there is still a paucity of data on some topics, non-peer reviewed sources or oral communication were also used when it could be corroborated by multiple sources.

All units of heavy metal and organic pollutant concentrations were converted to mg/kg = ppm for consistency and more straightforward comparisons. Some of the publications used for this synthesis reference other data sets but the reference material is unavailable. To support reliability, only the primary data sources were reported. The maximum concentrations that were detected in at least one individual fish from the family are reported here (Table 1.1).

Fish Sample Collection and Data Analyses

Sample Collection

Over the course of 12 months (2017-2018), 77 locally caught fishes representing 12 different taxonomic Families were purchased directly from roadside stands or from local stores in American Samoa. All fish were fresh (never frozen) and were generally caught the day they were acquired. Among the purchased fishes, reef and bottom dwelling species were collected using on-shore fishing gear or from small near-shore fishing vessels, and pelagic species were caught using local long-line vessels.

Heavy Metal Contaminant Analyses in Fish Muscle Tissue

The 77 fishes were subsampled for analyses of 41 different elements by Quadrupole ICP-MS (ThermoFisher Scientific iCAP Q, with CCT option). Approximately 0.2g of fish muscle tissue was collected and digested in reverse aqua regia (1 part trace metal grade 12M hydrochloric acid and three parts trace metal grade 15.6 M nitric acid) on a 120°C hotplate for 24 hours. Following the digestion period, each sample was then diluted up to a volume of 5 mL with nitric acid. Mercury (Hg) concentration was measured separately based on US EPA method 1631E, with on-line Hg(0) cold vapor generation by $SnCl_2$ reduction. Maximum concentrations detected in fish samples are reported in µg of metal contaminant per gram of wet tissue weight (e.g. parts per million or ppm). Analyses were conducted by the Keck Laboratory at Arizona State University.

Persistent Organic Pollutants Analyses in Fish Muscle Tissue

Following previously published methods (Polidoro et al. 2022, Pulford et al. 2017, Lucas and Polidoro 2019), each of the 77 fishes collected, a 5g sample of muscle tissue was extracted and spiked with 60 µg of p-terphenyl as a recovery surrogate, and then homogenized in 20g of Na_2SO_4 to remove excess water. Homogenized samples were then spun on a rotor for 48 hours in 60ml of 1:1 hexane:acetone. Solvent extracts were decanted and passed through several cleanup columns to remove larger molecules and polar compounds (e.g. Biobeads SX-3, Acidified Silica Gel, and Florisil). Sample extracts were then concentrated with nitrogen gas to a final volume of 0.5ml and spiked with tetracosane as an internal standard. All samples were analyzed for organic contaminants using a using a Varian 3800 gas chromatograph in tandem with a Saturn 2200 electron ionization mass spectrometer. Maximum concentrations detected in samples are reported in µg of organic contaminant per gram of wet tissue weight (e.g. parts per million or ppm). To estimate method recoveries, samples from yellowfin tuna were spiked with known concentrations of pesticides, phthalates, and PAHs. Method recoveries ranged from 30% to 50% for PCBs, 30% to 60% for pesticides, from 20% to 40% for phthalates, and from 20% to 90% for PAHs. Results presented are uncorrected for method recoveries, providing a conservative estimate of actual tissue concentrations. Analyses were conducted by the Polidoro Laboratory at Arizona State University.

Estimation of Fish Consumption Risk

The EPA has established oral reference doses for 10 of the metals reported by the reviewed literature (US-EPA IRIS database 2023). The oral reference dose is an estimate of the maximum recommended human daily oral exposure, below which there is unlikely to be risk of adverse effects over a lifetime. Oral reference doses are calculated based on assumed chronic consumption by an individual of average weight on a regular basis over a long-term period, and not set on the premise of one-time consumption. There is limited published data regarding Samoan body weight, this study estimates that the average American Samoan weight is approximately 100 kg (Pawson and James 1981), but this may be a low estimate. In order to directly compare the oral reference dose, which represents maximum recommended human exposures, with our metal and organic contaminant results reported in ppm, the oral reference dose can be used to create an Action Level. The Action Level is a screening-level threshold corresponding to the maximum contaminant concentration in a fish that is recommended to avoid long-term chronic impacts, based on average consumption rates and average human body weights (US-EPA 1998). It is calculated as follows:

Action Level (ppm) = (oral reference dose in mg kg-1 body weight x body weight (kg))/(serving size (kg) x # servings per week/ days per week).

Survey of fish consumption rates

This study is the first known to survey on island American Samoans to assess fish consumption rates. Most studies use consumption data based on mainland Americans, but

Samoans tend to consume more fish than mainland Americans. One record was found in which Pacific Islanders residing in San Francisco were estimated to consume 32 grams of fish per day (US-EPA 2006) (which translates to 0.224 kilograms per week, less than 1/3 of what my survey respondents reported), but the authors recognize that this estimation might not translate to people living on island. To more precisely calculate action levels, 37 adult individuals of Samoan decent were surveyed to assess their fish consumption rates. Of the 37 adults, 33 (89%) reported consuming fish. Most respondents reported eating 2-3 servings of fish per week, with some reporting significantly more. Therefore, Action Levels (in ppm) were calculated based on an average adult body weight of 100kg (Pawson and Janes 1981) and a serving of 0.3kg of fish consumed 3 times per week.

Results

Heavy Metals and Persistent Organic Pollutants in Fish from American Samoa: A Review of the Available Literature

There are a total of five official research studies that have analyzed and published data for heavy metal and persistent organic pollutants in commonly caught and consumed fish from American Samoa (AS-EPA 1994, Government of American Samoa 1991, Morrison et al. 2015, Peshut and Brooks 2005, US-EPA 2006), three from government agencies and two from peer reviewed scientific publications.

Fish acquire metals from the food they consume, the water they spend time in, and the sediments they encounter (Panda et al. 2023). The first study to evaluate potential contaminants in fish tissue in American Samoa was contracted by local government and non-government agencies in 1991 to create a baseline assessment of contaminants in fish muscle tissue (AS-EPA 1994). Fish collected from within the harbor were analyzed for heavy metals, pesticides, polychlorinated biphenyls (PCBs), and polycyclic aromatic hydrocarbons (PAHs). Criteria for fish collection required that they were commonly caught and consumed, that they were likely to spend most or all their life history within the harbor, and that they represented groups with different diets, including detritivores (Family: Mugilidae), herbivores (Family: Acanthuridae), and carnivores (Family: Carangidae). Although they weren't the main focus of the study, additionally more transient species from the family Lutjanidae were analyzed.

The study reported measurable concentrations of the heavy metals silver, arsenic, cadmium, chromium, copper, mercury, nickel, lead, and zinc in all fish samples. The metal in highest concentrations was lead, which had a maximum concentration of 7.9 ppm in an individual Mugilidae (Table 1.1), which might be indicative of elevated lead levels in the Harbor. Heavy metals were present in all fish samples (Table 1.1), with arsenic and mercury being in high enough concentrations from fish caught from the inner harbor that the EPA issued an advisory suggesting against consuming any fish caught within the harbor (AS – EPA 1999). Individual PCBs were not measured, but Aroclor PCB cocktails were measured and were undetectable in fish muscle tissue; Aroclor 1260 was present in the liver of two individual fish with a maximum concentration of 0.44 ppm. One individual fish had measurable concentrations of DDD and DDE, residues of the pesticide DDT (Table 1.2). No PAHs were detected in any of the fish tissue. The laboratory methods used for this study are undescribed (The Government of American Samoa 1991).

In a 1994 follow up study, the metals and organochlorine compounds were measured in fish muscle tissue, from a total of 8 different fish families (Tables 1.1 and 1.2) collected from sites around the island of Tutuila, with a priority for species that were commonly caught and consumed by local people (AS-EPA 1994). Most of the contaminant measurements were in ranges generally considered safe for human consumption, with the potential exceptions of arsenic and mercury. Total arsenic levels appear that they possibly had significantly increased since the 1991 study, which could have been a result of increases in arsenic pollution, but it's also possible there were variations in laboratory techniques or other unknown variables. It is also noticeable that had wide range of total arsenic concentrations were from the maximum concentration of 0.67 ppm in an individual in the Carangidae family to 21.3 ppm in and individual in the Holocentridae family, a range that makes it challenging to generalize.

Based on the results from AS-EPA (1994) and The Government of American Samoa (1991) studies, local authorities and researchers chose to focus only on the contaminants they deemed to have the most potential to cause negative health outcomes particularly in Pago Pago Harbor, which included total arsenic, mercury, lead, and PCBs (Peshut and Brooks 2005). There are distinct toxicity differences between the inorganic and organic fractions of total arsenic, but at this time the fractionation was poorly understood, therefore Peshut and Brooks (2005) assumed that the inorganic fraction of arsenic was 10% in fish tissue and reported a maximum converted concentration of 0.25 ppm inorganic arsenic, which is below the maximum recommendation estimated by the US-EPA Arsenic (2000) of 1.2 ppm. The conclusions of this study suggested that consumption advisories against consuming fish caught within Pago Pago Harbor were still supported based on (inorganic) arsenic, mercury, lead, and summed Arochlor PCB concentrations, but that fish caught elsewhere around the island were likely generally safe to consume (Peshut and Brooka 2005). Total arsenic was not reported, nor was the conversion factor, therefore the inorganic arsenic fraction is not directly comparable to total arsenic measurements identified in other studies. The specific laboratory techniques used are unclear for this study.

To better define the organic and inorganic fractionation in fish tissue, Peshut et al. (2007) conducted a follow up study in American Samoa which described shellfish of having higher fractions of inorganic arsenic than finfish, 1-5% and 0.5%, respectively.and thus, they estimate that approximately 99.5% of arsenic found in finned fish is in the organic form and is considered to result in minimal toxicity.

The earliest studies only reported measurable persistent organic pollutants in the form of the Arochlor PCB mixtures, but the two most recently published studies have identified persistent organic pollutants (other than Arochlor PCB mixtures) in measurable concentrations in fish tissue (Peshut and Brooks 2005, US-EPA 2006). One individual fish had measurable concentrations of DDD and DDE, and all fish families sampled had measurable concentrations of summed PCBs.

In 2006, the AS – EPA conducted a follow up study to assess mercury, arsenic, and PCBs in Pago Pago Harbor. They estimate that inorganic arsenic levels were below levels of concern, and there are still no known anthropogenic inputs into the harbor currently. They measured mercury concentrations for four families (Serranidae, Mullidae, Mugilidae, and Carangidae) of 0.15 ppm to 0.67 ppm, and summed PCB concentrations of 0.13 ppm to 0.64 ppm. The next and most recent published study of metals in fish was conducted in 2015 to better define the fractionation of methylmercury (from total mercury) in fish tissue (Morrison et al. 2015). Fish from the families Acanthuridae and Mullidae were measured and resulted in maximum concentrations of 0.013 ppm to 0.36 ppm, respectively, and they identified recovery percentages of methylmercury of 40% to 100% among individual fish, and that methylmercury concentrations often increased with fish body mass (Morrison et al. 2015).

Table 1.1 Heavy Metals in Fish Muscle Tissue in Fish Caught in American Samoa. Historical data: Maximum Heavy Metal Concentrations Grouped by Fish Family

Fish muscle tissue-Metals **maximum** concentrations

Collection Year	Fish Families	n^n	Ag	As	Cd	Cr	Cu	Hg	Ni	Pb	Se	Zn
	# Action Level	→	NA	NA	2.3	2.3	NA	*0.078	16	3.9	3.9	233
[a]1990		n^n	Ag	As	Cd	Cr	Cu	Hg	Ni	Pb	Se	Zn
	Acanthuridae	9	0.35	0.03	0.20	1.9	5.62	0.06	1.9	3.6	-	9.3
	Carangidae	2	0.15	<0.01	0.33	5.5	0.05	0.05	3.1	1.9	-	23
	Lutjanidae	1	0.13	<0.01	0.37	4.6	0.08	*0.08	4.1	2.5	-	12.2
	Muglidae	13	0.6	0.15	0.5	21.7	9.83	0.02	8.1	7.9	-	18.6
[b]1992		n^n	Ag	As	Cd	Cr	Cu	Hg	Ni	Pb	Se	Zn
	Acanthuridae	13	-	3.9	-	2.1	3.7	*0.21	2.6	1.25	0.26	4.2
	Carangidae	3	-	0.67	-	0.64	1.38	*0.57	3.3	0.25	2.3	12.5
	Holocentridae	10	-	21.3	-	1.2	28.1	*0.37	4.4	0.63	0.95	128.6
	Mugilidae	2	-	1.3	-	2.1	1.6	0.06	1.9	2.5	0.29	35
	Mullidae	1	-	3.5	-	0.55	0.69	0.03	0.9	0.13	0.14	6.3
	Scaridae	11	-	1.7	-	1.3	15.6	*0.15	3	0.66	0.82	91.2
	Scombridae	2	-	0.85	-	0.23	0.6	*0.19	0.83	5.98	0.41	6.2
	Serranidae	1	-	2.1	-	0.24	0.8	*0.26	0.85	0.43	0.22	3.8
[c]2005		n^n	Ag	As	Cd	Cr	Cu	Hg	Ni	Pb	Se	Zn

Acanthuridae	-	-	-	-	-	-	*0.13	-	0.04	-	-
Carangidae	-	-	NA	-	-	-	*0.42	-	0.005	-	-
Holocentridae	-	-	NA	-	-	-	*0.62~	-	0.01	-	-
Mugilidae	-	-	NA	-	-	-	0.02	-	0.05	-	-
Mullidae	-	-	NA	-	-	-	*0.5	-	0.1	-	-
Scombridae	-		NA	-	-	-	*0.21	-	0.007	-	-
Serranidae	-	-	NA	-	-	-	*0.67	-	0.16	-	-
[d]2002 & 2005	n[n]	Ag	As	Cd	Cr	Cu	Hg	Ni	Pb	Se	Zn
Carangidae	24	-	2.5^	-	-	-	*0.21	-	-	-	-
Mugilidae	12	-	1.3^	-	-	-	*0.15	-	-	-	-
Mullidae	1	-	-	-	-	-	*0.4	-	-	-	-
Serranidae	1	-	-	-	-	-	*0.67	-	-	-	-
[e]2015	n[n]	Ag	As	Cd	Cr	Cu	Hg	Ni	Pb	Se	Zn
Acanthuridae	60	-	-	-	-	-	0.013	-	-	-	-
Mullidae	21	-	-	-	-	-	*0.36	-	-	-	-

Maximum concentrations detected in at least one fish from the given family

[n] n = sample size

^ Converted to maintain consistent units for arsenic concentration

All concentrations are measured in mg/ kg = ppm wet weight

Mercury is reported as total mercury

~ Potential outlier, most concentrations were magnitudes lower

Action Level is calculated for an adult Samoan of average weight (100kg) consuming 0.3kg tissue 3 times per week, using US-EPA established oral reference doses.

[a] Government of American Samoa (1991)

[b] AS-EPA (1994)

[c] Peshut and Brooks (2005) - date of sample collection is unclear

[d] US-EPA (2006)

[e] Morrison et al. (2015) – date of sample collection is unclear

[*] Exceeded calculated action levels (based on calculations of current study)

- (dashed line) indicates the metal is not reported in the study

Table 1.2 Mercury in Fish Muscle Tissue in Fish Caught in American Samoa. Historical data: Maximum Total Mercury heavy metal Concentrations Grouped by Fish Family.

Maximum total mercury concentrations by fish family

Fish Families

	Acanthuridae	Caranidae	Holocentridae	Lutjanidae	Mugilidae	Mullidae.	Scaridae.	Scombridae	Serranidae
[a]	0.06	0.05	-	0.08	0.02	-	-	-	-
[b]	0.21	0.57	0.37	-	0.06	0.03	0.15	0.19	0.26
[c]	0.13	0.42	0.62	-	0.02	0.5	-	0.21	0.67
[d]	-	0.21	-	-	0.15	0.4	-	-	0.67
[e]	0.013	-	-	-	-	0.36	-	-	-
Overall maximum concentrations									
	0.21	0.57	0.62	0.08	0.15	0.5	0.15	0.21	0.67

All concentrations are measured in mg/ kg = ppm wet weight
Mercury is reported as total mercury
~ Potential outlier, most concentrations were magnitudes lower
[a] Government of American Samoa (1991)
[b] AS-EPA (1994)
[c] Peshut and Brooks (2005)
[d] US-EPA (2006)
[e] Morrison et al. (2015)

Table 1.3 Persistent Organic Pollutants in in Fish Caught in American Samoa.
Historical data: Persistent Organic Pollutants Maximum Concentrations by Fish Family

Year		Fish muscle tissue-Maximum Persistent Organic Pollutant concentrations				
	Fish Families	n*	PCBs Arochlor mixtures	PAHs	Pesticides	Phthalates
[a] 1990		-	0.038		(n=1) DDD = 0.038 ppm DDE = 0.016 ppm	
[b] 2005						
	Carangidae	-	0.21			
	Scombridae	-	0.10			
	Mugilidae	-	0.32			
	Mullidae	-	0.64			
	Serranidae	-	0.52			
[c] 2002 & 2005						
	Serranidae	1	0.52			
	Mullidae	1	0.64			
	Mugilidae	6	0.13			
	Carangidae	12	0.37			

*n = sample size
all concentrations are measured in mg/ kg = ppm wet weight
[a] Government of American Samoa (1991)
[b] Peshut and Brooks (2005) - date of sample collection is unclear
[c] US-EPA (2006)

Heavy metals and persistent organic pollutants: Original Research

Measurable concentrations of heavy metals (Table 1.4) and persistent organic pollutants (Table 1.5) in all fish samples. Total mercury was the only metal to exceed calculated action levels (Table 1.4). At least one individual from the families Holocentridae, Labridae, Lethrinidae, Mullidae, Scaridae, Scombridae, and Serranidae exceeded calculated action levels

Table 1.4 Heavy Metals in Fish Muscle Tissue in Fish Caught in American Samoa.
Original Research: Maximum Heavy Metal Concentrations Grouped by Fish Family.

	Maximum concentrations in fish muscle tissue										
Fish Families	n^n	Ag	As	Cd	Cr	Cu	Hg	Ni	Pb	Se	Zn
Action Level	→	NA	NA	2.3	2.3	NA	*0.078	16	3.9	3.9	233
Collection Year 2017 & 2018	n^n	Ag	As	Cd	Cr	Cu	Hg	Ni	Pb	Se	Zn
Acanthuridae	7	-	1.2	0.009	0.98	0.46	0.008	0.17	0.02	0.14	5.3
Chanidae	3	-	12.5	0.02	0.05	0.06	0.056	0.007	0.004	0.55	6
Haemulidae	1	-	8.9	0.004	0.04	0.42	0.072	0.01	0.01	0.19	6.6
Holocentridae	8	-	12.5	0.06	0.12	0.7	*0.32	0.03	0.02	0.5	7
Labridae	2	-	17.1	0.004	0.1	0.17	*0.16	0	0.013	0.26	3.1
Lethrinidae	5	-	18	0.03	0.06	0.14	*0.22	0.07	0.02	0.46	6.5
Lutjanidae	3	-	2.8	0.03	0.05	0.23	0.02	0.03	0.009	0.42	5.9
Mullidae	12	-	19	0.02	0.07	0.26	*0.22	0.13	0.03	0.52	4.1
Mugilidae	2	-	18	0.02	0.13	0.61	0.07	0	0.004	0.27	5.2
Scaridae	11	-	2.5	0.12	0.11	0.64	*0.13	0.37	0.01	0.22	6.2

Scombridae	16	-	7	0.01	0.09	2.3	*0.12	0.15	0.08	0.91	6.4
Serranidae	8	-	13.5	0.01	0.05	0.06	*0.23	0.05	0.009	0.47	4.6

Maximum concentrations detected in at least one fish from the given family

[n] n = sample size

^ Converted to maintain consistent units for arsenic concentration

All concentrations are measured in mg/ kg = ppm wet weight

Mercury is reported as total mercury

~ Potential outlier, most concentrations were magnitudes lower

[#] Action Level is calculated for an adult Samoan of average weight (100kg) consuming 0.3kg tissue 3 times per week, using US-EPA established oral reference doses.

[*] Exceeded calculated action levels (based on calculations of current study)

- (dashed line) indicates the metal is not reported in the study

Table 1.5

Persistent Organic Pollutants in in Fish Caught in American Samoa.

Original Research: Maximum Persistent Organic Pollutant Concentrations by Fish Family

		Maximum persistent organic pollutant concentrations in fish muscle tissue-			
		PCBs	PAHs	Pesticides	Phthalates
Fish Families					
Collection Year 2017 & 2018	n*				
Acanthuridae	7	0.07	3.1	0.3	0.6
Chanidae	3	0.05	0.26	0.01	2.3
Haemulidae	1	0.001	0.09	0.04	0.05
Holocentridae	8	0.02	0.82	2.6	2.4
Labridae	2	0.007	0.17	0.004	0.06
Lethrinidae	5	0.007	0.56	0.005	1.8
Lutjanidae	3	0.01	0.41	0.09	0.5
Mugilidae	2	0.001	0.13	0.005	0.02
Mullidae	12	0.04	0.55	1.6	1.1
Scaridae	11	0.03	7.7	0.9	2.7
Scombridae	16	0.01	1.5	0.7	0.5
Serranidae	8	0.03	0.88	0.08	3.1

*n = sample size

all concentrations are measured in mg/ kg = ppm wet weight

Discussion and Future Directions

Advances in technologies for sea travel advanced more people began landing in American Samoa, bringing along new technologies and foreign goods, which in turn brought as well as instances of land use change and environmental pollutants. The earliest identifiable records of pollution data in American Samoa were of people documenting visible environmental events, such as excessive algal blooms and fish die-offs. As technology and other analytical techniques were discovered and improved, and as people have become more aware of the negative environmental health outcomes, the data sets have been increasingly more robust. Heavy metals and persistent organic pollutants are found ubiquitously around the Island of Tutuila, which further impacts contaminant concentrations in the fish that reside and feed there, and subsequently the people that eat them. In parallel, fishing technology and effort has changed dramatically at the same time. While there have been enhancements in technology, the fishing industry has gone through booms and bust, and is currently in overall decline.

Including data from the current study, there are six total data sets related to heavy metal and persistent organic pollutants in commonly consumed locally caught fish from American Samoa. While several heavy metals were identified in the fish samples, mercury is the only heavy metal that has consistently been reported to be at concentrations potentially acutely hazardous for human consumption in American Samoa (Government of American Samoa 1991, AS-EPA 1994, Peshut and Brooks 2005, US-EPA 2006, Morrison et al. 2015, results from current study). Trends suggest that generally metals are not increasing in ways that warrant current concern, yet it is important to maintain periodical monitoring to ensure that the trends continue, particularly for mercury which can be toxic to humans who consume a lot of fish.

For the current study, we measured pollutant concentrations for 77 fish from 12 fish families; Acanthuridae (surgeonfish and unicorn fish), Scombridae (tunas), Holocentridae (squirrelfish), Scaridae (parrotfish), Serranidae (grouper and rockfish), Lethrinidae (emperor fish), Mullidae (goatfish), Lutjanidae (gray snapper, blue-striped perch), Labridae (wrasse), Chanidae (milkfish), Mugilidae (mullet), and Haemulidae (oriental sweetlips). Consistent with the previous five studies, the only contaminant that consistently exceeded calculated toxicity levels was mercury. The earliest reports of mercury in fish tissue from 1991 (Government of American Samoa 1991) reported a range of concentrations from 0.02 ppm to 0.08 ppm, with the highest concentration occurring in the family Lutjanidae. The follow up study by the AS -EPA (1994) expanded the number of diversity of fish and found a range of 0.03 ppm to 0.57 ppm, with the highest concentration occurring in the family Carangidae. In 2005, Peshut and Brooks identified a range of mercury concentrations from 0.02 to 0.67 ppm, with the highest concentration occurring in the family Serranidae. The follow up study from the US-EPA (2006) measured a range of mercury concentrations from 0.15 ppm to 0.67 ppm, with the highest concentration occurring again in the family Serranidae. In 2015, Morrison et al. (2015) measured a range of mercury concentrations from 0.013 ppm to 0.36 ppm, with the highest occurring for the family Mullidae (it should be noted that this study only sampled two fish families; Acanthuridae and Mullidae). The overall highest mercury concentration for the current study was 0.32 ppm in an individual in the family Holocentridae, with an overall range from 0.008 ppm to 0.32 ppm. The most recent study conducted by the AS - EPA (2006) hypothesize that persistent PCB and mercury concentrations might be caused from the Satala Power Plant and/ or Southwest Marine of Samoa, Inc. and recommend that these sites be subject to further monitoring. Results from the current study suggest that mercury might still be a potential toxicant of concern when consumed in excess from fish caught around American Samoa, but that the concentrations do not appear to be dramatically increasing or decreasing.

Overall lead concentrations were much lower than the first studies in 1991 and 1994 studies, which reported a maximum concentration of 7.9 ppm. Peshut and Brooks (2005) reported maximum concentrations of 0.1 ppm, and until the current study, lead in fish tissue had not been reported since 2005. Lead levels appear consistently lower in the current study than the earliest studies, which could indicate overall decreasing lead levels in fish or may be due to where the fish were collected. Because of the know high levels of pollutants (including lead) in Pago Pago Harbor, many of the previous studies focused on fish caught within the harbor, whereas the current study collected fish from around the island.

Most of the research of persistent organic pollutants in American Samoa has been on polychlorinated biphenyls (PCBs), particularly Arochlor mixtures. Even though the manufacturing of PCBs ceased in 1977 due to their elevated toxicity consequences, they continue to persist in the environment and transfer to fish. Regarding fish muscle tissue, only two studies from Peshut and Brooks (2005) and the US-EPA (2006) reported measurable concentrations of the sums of PCBs in American Samoa. In 1991, the only measurable pesticide was for the residues of DDT in one single fish (family: Mugilidae) with concentrations of 0.038 ppm DDD and 0.016 ppm DDE.

Based on the current study (Table 1.5) and ongoing local sediment and water monitoring, there appears to be more persistent organic pollutants being detected than have been previously reported, therefore it is suggested that persistent organic pollutants are periodically assessed in regularly consumed fish muscle tissue.

The trends in heavy metal and persistent organic pollutants in fish in American Samoa are challenging to generalize. Methods or variation in laboratory techniques are known to impact the detected concentrations between different studies, and further sometimes the sample size is too small to estimate trends for some contaminants. A greater diversity of organic pollutants was identified in the current study, which could be for multiple reasons, including there being more organic pollutants released into the environment, as well as more advanced analytical techniques. Regardless of why, ongoing monitoring of organic pollutants will provide relevant insight into changes over time. Due to the persistent presence of many heavy metals and persistent organic pollutants in fish, and because mercury continues to result in concentrations in fish muscle tissue that might lead to negative health outcomes, it is recommended that periodic and ongoing monitoring be maintained.

While there has been a decline in fishing effort, largely due to a shift from a subsistence lifestyle, there is no direct evidence that the shift has been due directly or indirectly to pollutants in fish tissue. In 1995, Tulagi and Green (1995) conducted a survey of public perception of the status of fishing resources and found that there was a general consensus that commonly consumed seafood abundance had been decreasing around the island, and some practices had nearly or completely ceased due to habitat destruction for the fish and other organisms, land use change, and pollution. Reduction in fishery resources due to overfishing is likely another reason for reduction in fishing effort (Craig et al. 1999), but it is unclear if the results of overfishing disproportionately contribute to decreases in fish consumption.

Among of the main challenges of this and the previous studies is identifying the source of pollutants in fish tissue. Mercury, which is historically and currently the primary metal of concern, can be a result of long-term atmospheric transport, making the source of mercury unclear. It is also challenging to make direct comparisons between the results of this study and previous studies due to variation of locations where fish were caught and overall fish movement, as well as potential differences between laboratory analytical techniques.

Although there are still major pollution challenges in American Samoa, progress has been made in recent years to educate local people and to mitigate the problems. In addition to continuing to conduct and support research, local government and non-government agencies have been focusing on promoting efforts to encourage the public to participate in the greater efforts to reduce waste and support greater environmental health. In 2014, the Director of the AS-EPA issued a public notice that they would start issuing fines to individuals and groups unlawfully purchasing, distributing, or using unlawful pesticides.

In response to the scientific studies indicating potentially toxic levels of pollutants, the government and non-government agencies have expanded research studies and have established guidelines to help maintain current healthy populations and to mitigate problems where possible (US-EPA 1991, 2023). Local programs, specifically through the school systems, have been educating students on the hazards of local pollution and how important it is to keep their island healthy and beautiful, and are resulting in positive outcomes. In 2022, the AS-EPA hosted a weeklong (Environmental Health) online class for high school students that also resulted in 2,000 pounds of electronic waste being collected by the Department of Education for recycling, instead of being sent to the landfill (AS-EPA 2022).

Sometime in the 2000’s American Samoa decided to ban the use of traditional plastic bags and replaced them with Biobags which had claims of being biodegradable. Unfortunately, it was not the successful solution that they hoped it would be. To decompose, Biobags need to have access to oxygen, which tends to not be available in landfills. When they decompose, Biobags break down into small plastic fragments, which then contribute to the microplastic pollution problem and can be hazardous for biota who might consume it.

One of the most successful campaigns to reduce waste has been the “Keep American Samoa beautiful” campaign which was developed by the AS-EPA in 2012 in response to visible trash problems island wide. Local people who adopt sections along the streets, streams, and the coast are provided gloves and garbage bags to help collect and reduce waste. The campaign is widely advertised and has resulted in visibly less trash in some common areas in just a few years (personal observation and personal communications). That said it is still common to see people throw garbage into streams and waterways. The solutions are not simple but reducing physical waste will also likely reduce some of the chemical waste.

While many efforts in American Samoa are underway to lead to healthier environments, with fewer physical and chemical pollutants, this remains a challenging task. Partnerships of government, non-government agencies, along with scientists, educators, and the next generations are critical in supporting an American Samoa with less pollution and waste.

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CHAPTER 2

HUMAN HEALTH CONSUMPTION RISKS FOR HEAVY METALS AND PERSISTENT ORGANIC POLLUTANTS IN LOCALLY CAUGHT FISH FROM AMERICAN SAMOA

Abstract

Persistent organic pollutants and heavy metals affect coastal communities world-wide, particularly for those who rely on coastal seafood as a major food source. For this study we evaluated persistent organic pollutants (POPs) and heavy metals (HMs) concentrations in the muscle tissue of 77 (plus one more for just heavy metals) commonly consumed fish caught coastally in American Samoa, which were divided into four groups according on their trophic- based dietary preferences. To establish a human health baseline risk assessment, action levels were then calculated based on oral reference doses where they have been established. Measurable concentrations of POPs and HMs were identified in the muscle tissue for every sampled fish. Only one POP, the pesticide *trans*-nonachlor exceeded calculated action levels, with no significant difference between fishes with different dietary preferences. Of the HMs with established oral reference doses only Mercury (Hg) exceeded calculated action levels. While the results of this study don’t warrant elevated concern, it is suggested that ongoing monitoring occurs to determine any significant change in local contaminants and thus potential health risks. Also, due to the lack of established reference doses for many of the contaminants detected, and unknown synergistic effects, the analyses should be revisited as the toxicity databases are updated.

Introduction

Fish consumption is a primary source of human exposure to heavy metals (HMs) and persistent organic pollutant (POPs) (Bosch et al. 2015, Schukla et al. 2022, WHO 2023). As a result of natural geological weathering and human activities, HMs such as mercury, arsenic and cadmium, and POPs such as pesticides, polychlorinated biphenyls and phthalates have been identified in coastal systems and fish tissue worldwide, sometimes at elevated levels that might present risks to humans (US-EPA/ FDA 2023, Karimi et al. 2012). Due to their potential harmful impacts to human health, regulations have been placed on the use and discharge of many metal and organic compounds (Stockholm Convention 2019, (US – EPA 2023), but many of these chemicals resist degradation and thus persist in the environment for years, accumulating through the food chain. In the environment, HM and POP concentrations widely vary by geographical location (Bonito et al. 2016, Karimi et al. 2012). Therefore, local monitoring of fish populations that are being eaten is critical to identifying more precise risk of impacts to human populations. Assessment of chemical contaminants in locally caught fish can identify consumption levels that might be hazardous to specific consumers and to inform polices that are committed to protecting public health (Karimi et al. 2012, Lucas and Polidoro 2019, Pulford et al. 2017).

American Samoa is a small (~200 km^2) remote island in the South Pacific with a rich history as a fishing community, relying on locally caught seafood as a primary food source (Levine and Allen 2009). Most fish are caught using recreational and small-scale commercial fishing vessels (AS Creel Survey 2011; Craig et al. 1999). In general, reef and bottom fishes are taken home for personal consumption, as well as sold to local stores and roadside stands. Pelagic fishes including tunas require larger boats and more fishing gear and are sold to the (on island) Starkist cannery, to local restaurants, and in roadside stands.

In American Samoa, land-based sources of contaminants include industrial and agricultural pollutants that can enter the near shore marine environment from discharge into rivers and coastal waterways (Craig et al. 1999, Polidoro et al. 2017). Water quality and fish contaminant levels are often related, as pollutants in fishes can be reflective of both past and current environmental contaminant concentrations (US-EPA 2006). Even after the use of chemical contaminants have ceased, many of them persist in the environment (Bonito et al. 2016, Craig et al. 1999, Mason and Whitall 2019). Due to the hydrophobic nature of many persistent organic pollutants, in the marine environment they tend to bind to sediments and organic matter, where they are then consumed by fish and other organisms (Agah et al. 2006, Honda and Suzuki 2020).

Local pollution sources in AS include leachate from the single unlined landfill, industrial chemicals, military waste, agriculture pesticide use, and intentional dumping (Polidoro et al. 2017). Due to potentially hazardous levels of arsenic, mercury, and PCBs in fish caught in Pago Harbor, the American Samoa Government issued fish consumption warnings in October 1991 (US-EPA 2006), which have not been officially lifted since then. More recent studies have identified pesticides (including DDT), mercury (at potentially toxic levels), as well as PBDEs, PCBs and phthalates in fish collected from American Samoa and surrounding waters (Bonito et al 2016, Mason and Whitall 2019, Polidoro et al. 2017, Whitall and Holst 2015). There is evidence that high consumption of fish contaminated with these compounds can lead to adverse health outcomes (Jiang et al.

2005). However, because contaminant concentrations vary by fish species and by geographic location (Karimi et al. 2012), predicting contaminant concentrations is challenging, and can be subject to small scale variability. Therefore, the primary objective of this study is to create a relevant human health risk assessment of fish consumption for native Samoans. Using a human health risk assessment framework (EPA 2014), consumption risk of organic contaminant and heavy metal concentrations detected in a variety of locally caught fishes in American Samoa were compared to calculated action levels to estimate risk. It is hypothesized that mercury will exhibit trophic biomagnification, and that PCBs and arsenic might be present in elevated concentrations based on previous studies conducted in American Samoa (Peshut 2005).

Materials and methods

Fish Sample Collection

Over the course of 12 months (2017-2018), 77 locally caught fishes representing 12 different taxonomic Families were purchased directly from roadside stands or from local stores in American Samoa. All fish were fresh (never frozen) and were generally caught the day they were acquired. Among the purchased fishes, reef and bottom dwelling species were collected using on-shore fishing gear or from small near-shore fishing vessels and can be classified by their primary dietary preferences (Figure 2.1): herbivorous (group A, Families Acanthuridae, Scaridae), plankton and small invertebrates (group B, Families Holocentridae, Chanidae, Mugilidae), and microbenthic organisms and small fishes (group C, Families Serranidae, Lethrinidae, Mullidae, Lutjanidae, Labridae, Haemulidae). Pelagic fishes (Family Scombridae (Image 2.1) caught from local, off-shore long-line fishing vessels (FRMD 2019), were classified separately (group D).

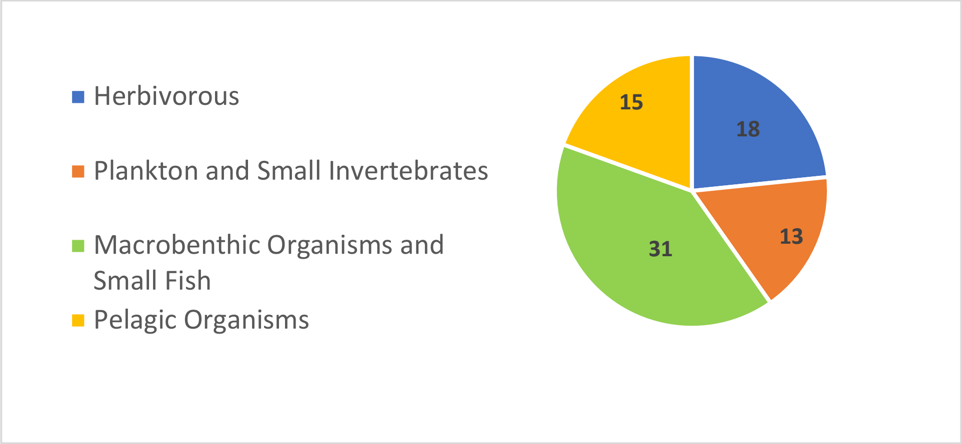


Figure 2.1 Four categories and number of individual fishes collected based on dietary preferences. Note that the subdivision may be imperfect due to some overlap in diets. N = 77

Figure 2.2 – Tuna samples. Fisherman Who Caught Some of the Pelagic Species (Images taken by Tiffany Lewis 2018)

Laboratory Organic Contaminant Analyses

Following previously published methods (Polidoro et al. 2022, Pulford et al. 2017, Lucas and Polidoro 2019), for each of the 77 fishes collected, a 5g sample of muscle tissue was extracted and spiked with 60 µg of p-terphenyl as a recovery surrogate, and then homogenized in 20g of Na_2SO_4 to remove excess water. Homogenized samples were then spun on a rotor for 48 hours in 60ml of 1:1 hexane:acetone. Solvent extracts were decanted and passed through several cleanup columns to remove larger molecules and polar compounds (e.g. Biobeads SX-3, Acidified Silica Gel, and Florisil). Sample extracts were then concentrated with nitrogen gas to a final volume of 0.5ml and spiked with tetracosane as an internal standard. All samples were analyzed for organic contaminants using a using a Varian 3800 gas chromatograph in tandem with a Saturn 2200 electron ionization mass spectrometer. Maximum concentrations detected in samples are reported in µg of organic contaminant per gram of wet tissue weight (e.g. parts per million or ppm). To estimate method recoveries, samples from yellowfin tuna were spiked with known concentrations of pesticides, phthalates, and PAHs. Method recoveries ranged from 30% to 50% for PCBs, 30% to 60% for pesticides, from 20% to 40% for phthalates, and from 20% to 90% for PAHs. Results presented are uncorrected for method recoveries, providing a conservative estimate of actual tissue concentrations. Laboratory analyses were conducted in the Polidoro Laboratory at Arizona State University.

Laboratory Heavy Metal Analyses

The 77 fishes were also subsampled for analyses of 41 different elements by Quadrupole ICP-MS (ThermoFisher Scientific iCAP Q, with CCT option). For each fish, approximately 0.2 g of wet weight tissue was collected and digested in reverse aqua regia (1 part trace metal grade 12 M hydrochloric acid and three parts trace metal grade 15.6 M nitric acid) on a 120°C hotplate for 24 hours. Following the digestion period, each sample was then diluted up to a volume of 5 mL with nitric acid. Mercury (Hg) concentration was measured separately based on US EPA method 1631E, with on-line Hg(0) cold vapor generation by $SnCl_2$ reduction. Maximum concentrations detected in fish samples are reported in μg of metal contaminant per gram of wet tissue weight (i.e., parts per million or ppm). Laboratory analyses were conducted in the Keck Laboratory at Arizona State University.

Estimation of Human Health Risk From Consumption of Contaminated Fish

For some, but not all, of the metal and organic contaminants detected, the EPA has established an oral reference dose (US-EPA IRIS database 2023). The oral reference dose is an estimate of the maximum recommended human daily oral exposure, below which there is unlikely to be risk of adverse effects over a lifetime. Oral reference doses are calculated based on assumed chronic consumption by an adult of average height and weight on a regular basis over a long-term period, and not set on the premise of one-time consumption. In order to directly compare the oral reference dose, which represents maximum recommended human exposures, with our metal and organic contaminant results reported in ppm, the oral reference dose can be used to create an Action Level.

The Action Level is a screening-level threshold corresponding to the maximum contaminant concentration in a fish that is recommended to avoid long-term chronic impacts, based on average consumption rates and average human body weights (US-EPA 1998). It is calculated as follows:

Action Level (ppm) = (oral reference dose in mg kg-1 body weight x body weight (kg))/(serving size (kg) x # servings per week/ days per week).

Survey of Fish Consumption Rates by Humans

This study is the first known to survey on island American Samoans to assess fish consumption rates. Most studies use consumption data based on mainland Americans, but Samoans tend to consume more fish than mainland Americans. One record was found in which Pacific Islanders residing in San Francisco were estimated to consume 32 grams of fish per day (US-EPA 2006) (which translates to 0.224 kilograms per week, less than 1/3 of what my survey respondents reported), but the authors recognize that this estimation might not translate to people living on island. To more precisely calculate action levels, 37 adult individuals of Samoan decent were surveyed to assess their fish consumption rates. Of the 37 adults, 33 (89%) reported consuming fish. Most respondents reported eating 2-3 servings of fish per week, with a few reporting significantly more. Therefore, Action Levels (in ppm) were calculated based on an average adult body weight of 100kg (Pawson and Janes 1981) and a serving of 0.3kg of fish consumed 3 times per week.

Statistical Analyses

To determine any patterns or trends in organic pollutant or heavy metal concentrations among fishes from different dietary groups, the concentrations of each heavy metal were compared between diet groups A-D. Because of the many detected PCBs, pesticides, phthalates, and PAHs, these categories were separately summed and then compared between diet groups. Due to the small sample size, and non-parametric nature of the data, Kruskal – Wallis statistical tests were used for these analyses. Dunn multiple comparison post- hoc test were conducted to further identify differences.

Results

Persistent Organic Pollutant Concentrations in Fish Muscle Tissue

Of the measured polycyclic aromatic hydrocarbons (PAHs), polychlorinated biphenyls (PCBs), pesticides, and phthalates, 13 have oral reference doses, including anthracene, biphenyl, carbaryl, DDT, DEPH, dibutyl phthalate, diethyl phthalate, hexachlorobenzene, methoxalchlor, 1-methyl (naphthalene), mirex, and nonachlor/ chlorodane (Supplementary Table 1). Measurable concentrations of POPs were detected in all fish groups but were all below calculated action levels except for one individual squirrelfish (Group B; family: Holocentridae) which had measured concentrations of the pesticide (*trans*-nonachlor) that far exceeded Action Levels (Supplementary Table 1). The Kruskal-Wallis tests identified no significant differences among any fish dietary groups for any contaminant class (Supplementary Table 2). This suggests that sampled fishes have similar risk for adverse health impacts based on these analyses of detected organic contaminant concentrations at this time, when consumed on average in 0.3 kg servings 3 times per week by Samoans with average body weights of 100kg. All non-significant statistical and graphical analyses are in the supplementary material under: Non-significant graphical analyses.

Polycyclic Aromatic Hydrocarbons (PAHs)

None of the samples analyzed for PAHs in this study exceeded calculated action levels. Of the 16 polyaromatic hydrocarbon (PAH) compounds identified across all fishes, only 2 have an established oral reference dose, anthracene and 1 – methylnaphthalalene. The highest concentration of anthracene was 0.0682 ppm found in an individual goatfish (habitat: reef; family: Mullidae), which is well below the calculated action level of 233.3 ppm. The highest concentration of 1 – methylnaphthalalene was 0.1172 ppm found in an individual grouper (habitat: reef; family: Serranidae), which is also well below the action level of 15.5 ppm. The two highest PAH concentrations measured were found in an individual parrotfish (habitat: reef; family: Scaridae), with a maximum concentration of 3.0471 ppm of chrysene + triphenylene and an individual surgeonfish (habitat: reef; family: Acanthuridae), with a maximum concentration of 1.2056 ppm of phenathrene.

Polychlorinated Biphenyls (PCBs)

None of the samples analyzed for PCBs in this study exceeded calculated action levels. 33 polychlorinated biphenyls (PCBs) compounds were detected across all sampled fishes. The only PCB with an established oral reference dose is biphenyl, which was identified in all fish diet categories, with the highest maximum concentrations occurring in resident reef fishes. The highest measured concentration of biphenyl was 0.0268 ppm in an individual grouper (Group C; family; Serranidae), followed by 0.0185 ppm in an individual surgeonfish (Group A; family: Acanthuridae), then 0.0168 ppm in squirrelfish (Group B; family: Holocentridae). The highest maximum concentration among the pelagic fish species was identified in an individual Skipjack tuna (Group D; family: Scombridae) with a concentration of 0.0022 ppm. The maximum concentrations are far below the action level of 388.9 ppm.

Pesticides

One individual squirrelfish sample (Group B; family Holocentridae) had a measured *trans*nonachlor pesticide concentration of 1.343 ppm which exceeds the action level of 0.389 ppm of the parent compound chlordane. Of the 9 pesticides identified in this study, seven have a US-EPA established oral reference dose. Several banned pesticides were present in measurable concentrations, including DDT, methoxychlor, and mirex, none of which exceeded calculated action levels calculated for adult Samoans. Maximum concentrations of DDT were identified in Groups B and D at 0.0006 ppm and 0.1004 ppm respectively, with the highest concentration occurring in an individual Skipjack tuna (Group D; family Scombridae). DDD, a breakdown product of DDT was identified in all fish groups. Methoxychlor and mirex were also detected in all fish groups, with maximum concentrations of 0.9262 ppm found in Group A and 0.0401 ppm in Group C, respectively.

Phthalates

None of the samples analyzed for phthalates in this study exceeded calculated action levels. Seven different phthalate compounds were identified across all fishes sampled. The phthalate with the highest maximum concentrations was Di (2-ethylhexyl) phthalate (DEHP). The highest concentration of DEHP was 2.674 ppm in an individual grouper (habitat: reef; family: Serranidae), the second highest concentration of 2.2383 ppm occurring in an individual parrotfish (habitat: reef; family: Scaridae), and third highest concentration of 2.0576 ppm occurring in an individual squirrelfish (habitat: reef; family: Holocentridae). The highest concentration of DEHP in pelagic fishes was 0.2595 ppm. All the measured concentrations of DEHP were below the 15.5 ppm action level. In addition to DEHP, 2 of the 7 identified phthalates have oral references doses, dibutyl phthalate and diethyl phthalate, which were detected in maximum concentrations of 0.5083 ppm and 0.0212 ppm respectively, and both well below the associated action levels of 77.8 ppm and 622.2 respectively.

Heavy Metals Concentrations in Fish Tissue

Of the 41 measured elements, only 11 metals have established oral reference doses: aluminum (Al), arsenic (As), cadmium (Cd), chromium (Cr), mercury (Hg), manganese (Mn), nickel (Ni), lead (Pb), selenium (Se), uranium (U), zinc (Zn). Of these 11 elements, only the maximum detected concentrations of mercury (Hg) and arsenic (As) exceeded calculated action level limits for a 100 kg adult consuming 0.3 kg of fish three times per week (Tables 2.1 and 2.2). It should be noted that the oral reference doses and thus action levels for Hg and As are all based on a specific elemental forms with elevated toxicity, and should be considered contextually.

Table 2.1

Heavy Metals: Maximum Concentrations and Action Levels for Fish Muscle Tissue.

	Groups based on trophic -level dietary preferences				
	A Primarily herbivorous	B Plankton and small inverts.	C Macrobenthic organisms and small fish	D Pelagic organisms	Action Level
Metal	Maximum concentrations (ppm)				
Aluminum (Al)	15	8	46	14	778
Arsenic (As)	2.5	18	19	7	[a]0.23
Cadmium (Cd)	0.12	0.06	0.03	0.01	2.3
Chromium (Cr)	0.98	0.13	0.07	0.09	2.3
Copper (Cu)	0.64	0.7	0.26	2.3	NA
Iron (Fe)	6.8	15	7.3	13	NA
Mercury (Hg)	*0.13	*0.32	*0.23	*0.12	*0.078
Manganese (Mn)	0.03	0.03	0.12	0.03	109
Nickle (Ni)	0.37	0.03	0.13	0.15	16
Lead (Pb)	0.02	0.02	0.03	0.08	3.9
Selenium (Se)	0.22	0.55	0.52	0.91	3.9
Uranium (U)	0.0006	0.0004	0.0005	0.0004	2.3
Zinc (Zn)	6.2	7	6.6	6.4	233

mg/ kg wet weight = ppm

*The maximum mercury (Hg) concentration for all groups exceeded the calculated action level based on a 100 kg adult show consumes 0.3 kg servings of fish muscle tissue 3 times per week.

NA = oral reference dose not available, therefore action level cannot be calculated.

[a] The EPA reports an oral reference does for inorganic arsenic, which is toxic and incomparable the less toxic organic form typically found in fish tissue.

All Action Levels here are calculated from the EPA's Integrated Risk Information System (IRIS) oral reference dose database.

Similarly, Kruskal-Wallis tests showed significant differences (<0.05) between at least two dietary groups for aluminum, arsenic, cadmium, iron, mercury, lead, and zinc, which were further explored using the Dunn multiple comparison post- hoc test (Supplementary Table 4) (Note: Only the HMs that exceeded calculated action levels will be discussed further). The most prominent variation between groups is for arsenic, which exhibited significant differences in concentrations between all diet group combinations (Supplementary Table 4). Summary statistics for all metals can be found in Supplementary Tables 5a, 5b, and 5c.

Table 2.2
Mercury: Maximum Concentrations and Action Levels for Fish Muscle Tissue

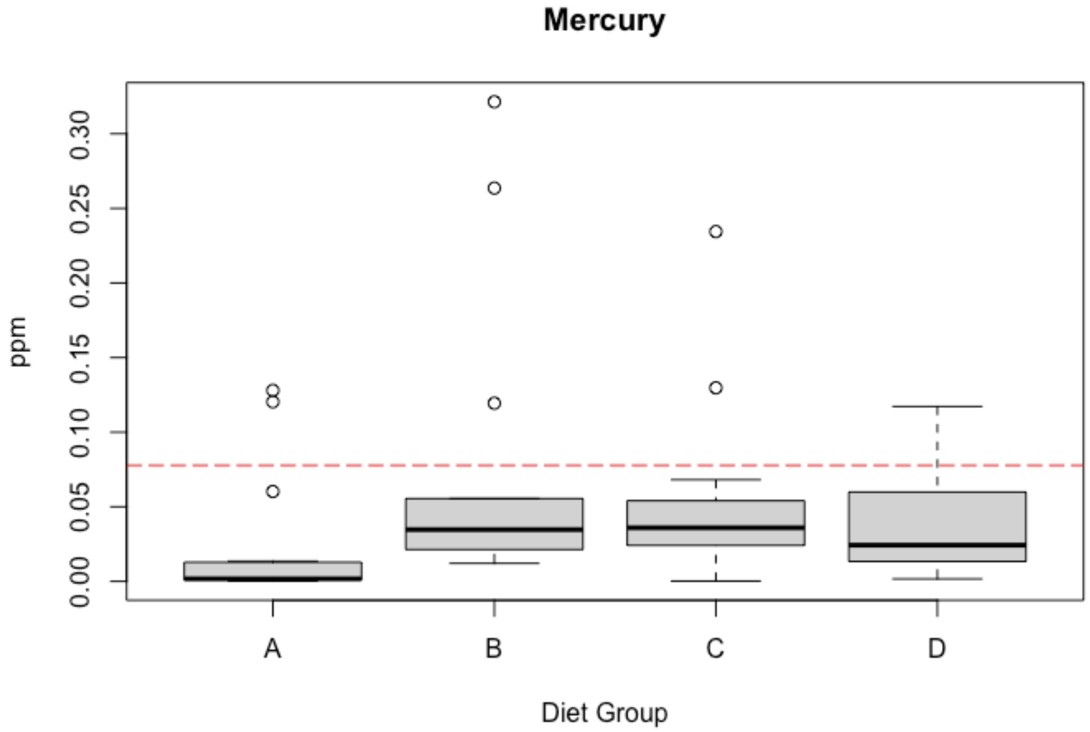
Element	Group A	Group B	Group C	Group D	Action Level
Mercury (Hg)	0.13	0.32	0.23	0.12	*0.078

mg/ kg wet weight = ppm
*The maximum concentrations from all trophic based diet groups exceeded calculated action levels

Mercury

Considering all fish, total mercury had a range of 0.0002 – 0.32 ppm and a mean of 0.04 (+/- 0.06). All of the maximum mercury concentrations identified in each fish group exceed calculated action levels of 0.078 ppm (Figure 2.3), but none of the means exceeded action levels. The highest concentration of mercury identified in this study (0.32 ppm) was in an individual squirrelfish (Group B; family: Holocentridae), and the lowest maximum concentrations among the groups was measured in the pelagic fishes (0.12 ppm).

Figure 2.3
Mercury: Relationship of Mercury Concentrations Between Trophic Based Dietary Groups



Kruskal - Wallis rank sum test comparing Hg concentrations by fish diet category. Kruskal – Wallis chi-squared = 16.7, df = 3, p-value = 0.0008 Dotted line indicates calculated action level (0.08 ppm).

Arsenic

Total arsenic concentrations were in the range of 0.28– 19.1 ppm, with a mean of 3.99 (+/-4.3) ppm. The highest maximum concentration of 19.1 ppm was identified in an individual goatfish (Group C; family: Mullidae). The US- EPA has an established oral reference dose for inorganic arsenic of 0.0003 mg/ kg/ day, which results in an action level of 0.23 ppm, yet fish mostly contain organic As, which exhibits significantly less toxicity (Taylor et al. 2017).

Discussion and Future Directions

This study provides one of the first relatively comprehensive baseline screening for heavy metal and organic contaminant concentrations in locally sourced seafood in American Samoa based on an 100kg adult consuming 0.3kg of fish muscle tissue 3 times per week. Any aberrations from this guideline might result in alternative risk, therefore, must be considered individually when choosing fish consumption; for example, residents with weights of less than 100kg or those who consume greater than 3 – 0.3kg of fish per week will have a calculated higher risk than those presented in this study. In addition, synergetic effects of multiple contaminants consumed at the same time are important to consider but difficult to quantify. It is also important to note that many contaminants detected do not have established oral reference doses, and there are also likely contaminants present in fishes that were not able to be detected with GCMS, such as pharmaceuticals, or are present in levels below method or instrumentation detection limits.

All fish samples had measurable concentrations of a variety of persistent organic pollutants and heavy metals. Most of the contaminants with established oral references were below calculated action levels for adult Samoans weighing 100kg and consuming 0.3kg of fish muscle tissue three times per week. The risk at the estimated consumption rates and body sizes appears to be highest for the pesticide *trans*nonachlor (from one individual fish), and the heavy metal mercury. However, it is important to note that many of the detected contaminants do not have established oral reference doses, and the synergistic effects are unknown, so this study is a conservative estimate of the actual risk.

Persons with increased consumption rates and decreased body sizes will have increased risk.

Also, there were no obvious patterns to contaminant distributions based on fish dietary groups, this potentially could be due to the opportunistic sampling method or lack of demographic data for the sampled fish, of which cannot be teased out for this study.

Persistent Organic Contaminants

PAHs occur naturally in the environment in crude oil, coal, and gasoline, and unnaturally as a common product of incomplete combustion primarily from human activities. Of the two highest PAH compounds measured, chrysene + triphenylene and phenanthrene are primarily derived from incomplete combustion from industrial activities, and chrysene is used to produce UV filters, paints, and fluorescent labeling. Chrysene + triphenylene have both been identified as mutagenic in laboratory tests, and chrysene is suspected to be carcinogenic and is one of 16 PAHs listed as a Priority Pollutant by the US EPA.

Along with Mercury, PCBs have been of historical concern for human health in American Samoa, particularly in Pago Harbor (Peshut et al. 2008). PCBs are a group of synthetic compounds used from 1929 for many commercial and industrial applications due to their chemical stability, until they were generally banned in 1979 by the Toxic Substances Control Act (TSCA). 33 PCB compounds were identified in this study. Although they have been banned for decades, PCBs can still be found in products produced before 1979 or in the environment where they persist bound to sediments and in organisms. Previous studies reported by the AS-EPA have reported measurable concentrations of the PCB mixtures Aroclors 1254 and 1260 but did not report individual PCBs (AS-EPA 1996, AS-EPA 2006). Due to the large number of individual PCBs and PCB mixtures, past concentrations are challenging to compare to the current study, and overall toxicity is unclear.

Only one pesticide in one individual fish exceeded calculated action levels. *Trans*nonachlor is one component of the pesticide chlordane, of which most studies have collected data toxicity for. Historically chlordane was primarily used for termite and other insect control from 1948 until it was banned in 1988 due to negative health effects, including neurological toxicity, digestive distress, liver damage, and in some cases convulsions and death. Other components of chlordane include cis- nonachlor, cis- chlordane, trans- chlordane, all of which were not detected in this individual fish. Chlorodane contains ~45 components of which *trans*nonachlor makes up approximately 15% of the total compound, depending on the formulation (US-EPA IRIS Database 1998). It is among the most common residues of the parent compound chlorodane found in the tissues of higher organisms, including fish and humans, and is among the most bioaccumulative of the chlorodane components (US-EPA 2004). One individual squirrelfish sample (Group B; family Holocentridae) had a measured *trans*nonachlor pesticide concentration of 1.343 ppm which exceeds the action level of 0.389 ppm of the parent compound chlordane. This elevated concentration is much higher than any other individual fish measured in this study and was well above the mean of 0.008 ppm, indicating that overall, it is unlikely that there are broad health risks associated specifically with *trans*nonachlor in AS at this time, but it is recommended that sampling and analyses be revisited in the future to reassess risk, and to determine if there more individuals with elevated levels.

Among the most frequently identified organic contaminants were phthalates, which generally are derived from the breakdown of plastic materials. The phthalate with the highest maximum concentrations was Di (2-ethylhexyl) phthalate (DEHP). DEHP was banned for use in children's products in 2018 due to risks associated with negative reproductive and developmental health outcomes. Although it is reasonable to hypothesize that one might measure higher concentrations of phthalates in reef fish, inhabiting and feeding in ecosystems with heavy plastic pollution, the individual variability among fishes restricts this study from forming these conclusions. While none of the individual phthalate concentrations did not exceed known action levels, cumulative effects were not measured and are currently unknown.

Heavy Metals

Mercury is the most frequently monitored contaminant in fish due to its potential toxicity to humans. In the early 1970s the first advisories for fish consumption in the United States were issued in response to elevated mercury levels in local waterways resulting from industrial discharge (Hesse 2005). The US Environmental Protection Agency (EPA)/ US Food and Drug Administration (FDA) generally suggest maximum fish consumption limits based a on metanalyses of select publications, but further recommend that local conditions are considered when choosing which and how much fish to eat. Currently, the FDA/ EPA only make specific consumption recommendations for pregnant or breastfeeding individuals and children of 8 to 12 oz. (0.23 – 0.34 kg) of low (methyl)mercury fish per week, and that children eat less than that based on their size. They calculate screening values for fish muscle tissue and classify them into 3 categories; choices to avoid, good choices, and best choices, largely dependent on mercury concentrations. The peer reviewed literature reports mercury concentrations in fish tissue to contain orders of magnitude variability among studies, and are divergent from EPA calculations, which might provide varying implications for public health (Karimi et al. 2012). Although mercury is a naturally occurring element, human exposure even in small amounts to methylmercury can be toxic, and toxicity can vary from person to person. It is among the top ten of major public health concerns by the World Health Organization (WHO 2023) due to its toxic effects on the nervous, immune, and digestive systems; the most severe cases resulting in neurological and behavioral disorders and death (Hong et al. 2012). Although there are natural sources of mercury, appreciable concentrations are often derived from human based industrial activities such as mining or burning coal (Hesse 2005).

Mercury enters the environment from natural processes such as volcanoes and fires, and anthropogenically through discharge from mining and other industrial activities. The toxic fraction of total mercury is in the form of methylmercury and constitutes up to 100% of total mercury in fish muscle tissue (Horvat 2005); Agah et al. 2007 identified the percentage of methylmercury in a range of 64% -100%, and Dezfouli et al. 2017 found that methylmercury constituted approximately 90% of total mercury in fish. Methylmercury is the most toxic form of mercury for humans and primarily enters the body from consuming contaminated fish (Harris et al. 2003; Hovart 2005).

The highest concentration of mercury identified in this study (0.32 ppm) was in an individual squirrelfish (Group B; family: Holocentridae), and the lowest maximum concentrations among the groups was measured in the pelagic fishes (0.12 ppm), which is contrary to trends that have been reported from previous studies exhibiting biomagnification; this results in the rejection of the hypothesis that mercury measurements in fish from this study follow established trends of trophic biomagnification. Concentrations in tuna and grouper species in this study were less than have been reported by FDA meta-analyses; this study identified 0.12 ppm maximum in fresh tuna and 0.07 ppm in fresh grouper, the FDA reports 1.8 ppm maximum in fresh/ frozen tuna 1.2 ppm maximum in grouper species. The US-EPA 2006 reported that the maximum mercury concentrations from fish collected from Pago harbor were 0.15 ppm for Mugilidae *spp*., 0.4 ppm for Mulidae *spp*, and 0.67 ppm for Serranidae *spp*., which were all higher than the measurements for this study which were 0.024 ppm, 0.23 ppm, and 0.07 ppm respectively. Although the measured concentrations of mercury exceeded calculated action levels in this study, the concentrations in fish muscle tissue from AS are lower than the widely available estimations reported by the EPA.

Mercury concentrations vary between species, between fish within a species, and by geographical location. Fish muscle tissue from commercially available fish were collected from a fish market in Kamamoto, Japan in 2012 and were analyzed for mercury concentrations with a range of 0.039 (0.02) ppm to 0.202 (0.12) ppm, with the highest concentrations occurring in Scorpion Fish, which are categorized as carnivorous, indicating that they are higher in food chain and might bio magnify for these groups (Watanabe et al. 2012). Four species including cod, eel, sprat, and herring were collected from the Baltic Sea in 2016 and analyzed for mercury, resulting in an overall range of 0.02 ppm in Baltic Sprat to 0.44 ppm in eel muscle tissue (Polak-Juszczak 2018). Because the majority of global mercury pollution is due to coal combustion used for global energy production, Sacket et al. 2010 assessed if proximity to coal fired plants contributed to the variation in mercury concentrations in fish, and determined that proximity to the source was not significant, but that fish variation (such as place in the food chain, weight, and age) contribute greater variation in totally mercury concentrations (Sacket et al. 2010). Scientists collected samples of consumable fish from around the Galapagos Marine Reserve, including five demersal species and four pelagic species, including *Thunnus albacores* (Albacore Tuna). The overall range of concentrations for all fish species were below detection in common dolphinfish and the highest overall concentration of 1.82 ppm in *Hyporthodus mystacinus* (Misty Grouper), and the range for just Albacore Tuna was 0.25 ppm to 1.62 ppm in muscle tissue (Franko-Fuentes 2023). The overall mercury results of the current study fall within the ranges, but below maximum concentrations identified in other published data, with an overall maximum concentration of 0.32 ppm from all sampled fishes. These individual studies reflect findings of metanalyses conducted by Karimi et al. 2016, which suggest that variation in mercury concentrations in fish muscle tissue varies based on species and location and must be considered in context when assessing true human health risk (Karimi et al. 2016).

Arsenic is a widely distributed element in the earth’s crust, with the most common human exposure resulting from anthropogenic sources such as mining, smelting, combustion, and pesticide application, and from natural sources such as volcanic materials, wind-blown soils, and naturally elevated and liberated ground deposits (EPA Arsenic 2006).

It enters the water and food supply from sediments and groundwater sources, combing with other substances to result in both inorganic and organic compounds. Inorganic arsenic compounds are associated with high toxicity, while organic arsenic is reported to be much less so. Among the most common sources of inorganic arsenic ingestion in humans is from drinking contaminated water, eating contaminated foods, from contact as a pesticide compound (EPA Arsenic 2006). The primary source of organic arsenic is from fish consumption, and is reported to have minimal negative health outcomes, although scientists are continuing research to better describe the effects of the organic fraction (Taylor et al. 2017). Arsenic concentrations exhibit high spatial variability, with specific communities being more at risk due to local contamination, and in elevated concentrations are carcinogenic (Khan et al. 2003).

There are no clear anthropogenic inputs of arsenic into coastal American Samoa, and at this time it is assumed that most of the arsenic in fish is a product of weathering and erosion of land based volcanic sediments, and minimally as a byproduct from industrial processes (US-EPA report 2006; CDC 2023). In comparing the highest total arsenic of 1.3 *ppm* in the US-EPA 200study to the highest concentration of *Mugilidae spp.,* we find significantly higher concentrations in the current study of 19.1 *ppm.* A study contracted by the US-EPA in 2006 measured 12 individuals each *Mugilidae spp.* collected from Pago harbor reported maximum concentrations of inorganic arsenic of 0.1298 ppm, by assuming that the inorganic partition was 10% of the total Arsenic concentrations.

Total arsenic was identified as a potential contaminant of concern in American Samoa (Peshut 2005), which prompted research to determine the inorganic and organic fractions in fish and shellfish in AS (Peshut et al. 2008). They determined that the fraction of inorganic Arsenic in fish tissue was generally less that 0.5% of total As, but that bivalves contained inorganic fractions up to 5%. Arsenic follows trends of biodilution, where lower trophic level organisms, such as bivalves tend to have higher concentrations of arsenic than mobile fish (CDC.gov 2022, Peshut et al. 2008).

The results of this study indicate that As exhibits the greatest variability in concentrations between trophic groups of all of the measured contaminants (Supplementary Table 3), with a mean of 4 ppm (total As) and the highest measured concentration of 19.1 (total As) ppm in an individual goatfish (Group C; family: Mullidae). The methods used in this study measures total inorganic and organic arsenic combined, therefore is not directly comparable to the US- EPA established oral reference dose of 0.0003 mg/ kg/ day for inorganic arsenic which results in an action level of 0.23 ppm. If it is assumed that the inorganic fraction is 0.5% (Peshut 2008), the highest calculated concentration is 0.96 ppm (inorganic As), which is still greater than the action level of 0.23 ppm but is below the mean adjusted concentration of 0.2 ppm (inorganic As).

Given the high variability and probable fractionation of toxic inorganic As, current arsenic concentrations might contribute to negative health outcomes in American Samoa, and ongoing monitoring is recommended to monitor As concentrations and fractionations, and as research into the effects of organic arsenic are further explored (Taylor et al. 2017).

This study focused on the maximum measured concentrations, identifying conservative a risk assessment (see Appendix for additional descriptive statistics). Based on the detected contaminants quantified in this study, fish in general do not appear to pose significant health risks to American Samoans based on average consumption rates and conservative risk calculations but given that there are individual fish with elevated contaminant concentrations and there is increasing identification of organic pollutants in fish tissue, it is important to maintain ongoing monitoring.

Future studies are recommended to include more information on the location at which fishes are caught to better relate detected contaminants to specific geographic areas or habitats, as well as to incorporate measurements of fish body size and estimated age. Measuring additional contaminants, such as pharmaceuticals, would also help to create a more comprehensive assessment of the risk of consuming fish. Finally, more detailed information on consumption rates for different human populations and preferred species would be useful, especially as tunas for example, are gear and cost prohibitive for many families, which may limit consumption.

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CHAPTER 3

FROM WILD CAUGHT TO CANNED: HUMAN HEALTH CONSUMPTION RISK OF HEAVY METALS FOUND IN COMMONLY CONSUMED CANNED MEATS IN AMERICAN SAMOA

Abstract

Canned corned beef, canned tuna, and fresh caught tuna are commonly consumed food items in American Samoa, with a dramatic shift from fresh local food to pre-packaged processed goods. With an expanding diet in Samoa, it is important to clarify contaminant concentrations in commonly consumed food items. This study measures and compares heavy metal concentrations among these popular food items, providing data and that allows people to make more informed choices about the foods that they consume.

89% of survey respondents reported consuming canned corned beef, and 89% of respondents also reported consuming canned tuna. 14 cans of commonly consumed (in American Samoa) canned corned beef and canned tuna with independent batch numbers were analyzed for heavy metal concentrations. The concentrations of the canned tuna and corned beef were compared to each other and then to fresh tuna. Action levels were calculated to assess risk of chronic consumption of these items. Results indicate that all metals in corned beef fell below calculated action levels, and only mercury (Hg) results were at high enough levels to exceed action levels of 0.078ppm, and fresh tuna had overall higher concentrations than canned tuna.

Introduction

Globalization and the introduction of novel food items has influenced dietary shifts from locally sourced foods to processed food items (Bindon 2006; Ichiho et al. 2013; Melby 2011). This shift has increased food based negative health outcomes, including increases in obesity and heart diseases, but there isn't locally available data regarding the heavy metal concentrations in commonly consumed canned tuna and corned beef in American Samoa. This study explores the differences in heavy metal concentrations between commonly consumed food items in American Samoa, canned corned beef, canned tuna, and fresh locally caught tuna.

Processed and prepackaged foods are a relatively novel development for populations world-wide, with the most prominent shift from locally grown or harvested food items to imported and pre-packaged foods occurring within the past approximately 200 years, and even more dramatically in the past 100 years (Melby 2011). The dietary shifts for the (American) Samoan population have occurred and been documented most prominently since the mid to late 20th century as the relationships particularly with mainland America, New Zealand, and Australia were enhanced and as global trade increased. Traditionally Samoans relied heavily on local food sources such as coastal fishes and shellfish, taro, coconuts (Figure 3.1), and sometimes chicken and pork, but in recent years that has shifted to include a higher proportion of imported and prepackaged foods.

Figure 3.1 Burning Coconut Shells into Coals For Cooking. Taro Root in Fresh Coconut Milk and Onions (fa'alifu kalo) Eaten on a Banana Leaf Plate (Images Taken by Tiffany Lewis 2018)

Early imports of processed canned foods products were supplied to American Samoa to supplement their food supply, particularly protein. Around the mid 20th century, the United States and New Zealand began sending turkey tails and mutton flaps (respectively) to Pacific Island nations, which are animal parts deemed undesirable for human (and often even for pet) consumption in their countries of origin (BBC 2016; Errington and Gewertz 2008). After World War II, industrialized turkey production was rapidly increasing in the US, creating an excess of turkey tails (an oil filled gland at thease of a turkey, which is ~75% fat), which until then was considered a waste product (Smithsonian Magazine 2017). Similarly, mutton flaps (the traditionally undesirable fatty part of the lower sheep ribs) began being imported from New Zealand (Errington and

Gewertz 2008). At face value this appeared to be a win-win situation; more food for island nations and less food waste products for the countries providing them.

Turkey tails, mutton flaps, and canned meat products are all novel protein sources that have been incorporated into the American Samoan diet and culture (Bindon 2006). All these products are commonly consumed during celebrations and are considered an integral part of life. Canned meat products have become even more incorporated into everyday menus due to its high palatability and simplicity to prepare. Canned foods have a relatively long shelf life and are pre-cooked and can be consumed right out of the can. The canning process itself kills most microorganisms that might cause illness and the can maintains an oxygen free environment allowing for longer shelf life, which can be particularly helpful in times when fresh food is scarce (Zheng et al. 2021).

Among the most popular canned food items in American Samoa is canned corned beef, referred to as ‘pisupo’. The term originated from the first tinned food product to be imported from New Zealand, which was pea soup. Because Samoan words must end in a vowel, the word was modified to pea-soup-o, later becoming a general term for canned foods, and then ultimately a name specifically for canned corned beef. The introduction of canned foods has fundamentally changed the diet and culture of the Samoan population (Simes 2018).

In 2007, Samoa attempted to ban canned processed meats because they are known to lead to diabetes and heart disease, but this was met with negative feedback, and ultimately the World Trade Organization (WTO) required lifting the ban in 2013 as a condition of membership (Gewert and Errington 2007). Canned meats are commonly purchased for family consumption, whether warmed up or eaten straight out of the can. It is common to provide a case of large (11.5 oz) cans of corned beef (*pisupo)* as a gift at many types of celebrations (*fa'alavelave)*, from birthdays to funerals. When asked why they eat pisupo, one of my survey respondents claimed, "Pisupo is Life". Most people surveyed share similar feelings. In American Samoa it is generally accepted that a diet heavy in processed canned meats may contribute to undesired health outcomes and, but the desire to change habits to increase quality of life is currently minimal (Simes 2018).

While the invention of canned foods has positively impacted accessibility to essential calories for populations lacking access to food, it also has led to increases in negative health outcomes, particularly when consumed in excess. The perception of whether a food item is deemed healthy or not is dependent on an individual's understanding of complex nutritional concepts, potential toxicity of the food products, and their cultural and social perception of the foods (Bindon 2006). And while it has been established by medical professionals that consuming excessive amounts of canned meats contributes to human disease such as, the extent to which heavy metals might contribute and compare to more traditional protein choices is unclear for this population.

It is challenging to generalize risk for most populations due to the spatial variation in dietary preferences and food availability, yet due to its relatively low and isolated population, American Samoans have a more homogenous diet than does mainland America, and thus it is more straight forward to make comparisons between items they regularly consume.

The objectives of this study are to,

a) Measure and compare heavy metal concentrations for three commonly consumed items in American Samoa; canned corned beef canned tuna, and fresh caught tuna.

b) Calculate action levels for canned corned beef and canned tuna to create baseline human health risk assessments for all heavy metals that have established oral reference doses.

c) Statistically compare heavy metal concentrations between canned tuna and canned corned beef, and canned tuna and fresh tuna.

Materials and Methods

Canned Tuna and Canned Corned Beef Sample Collection

14 cans of Starkist tuna in oil (ingredients: light tuna, soybean oil, water, vegetable broth, salt) and 14 cans of canned corned beef (Ingredients: cooked beef, water, salt, sodium nitrate) from four regularly consumed brands (Ox and Palm, Palm, Pacific, and Crown)(Figure 3.2) were purchased from various international stores in Arizona, USA. Each can has a unique lot number, thus representing independent batches. All cans were analyzed within the sell by date. These samples were collected because they specifically represent the foods that American Samoan people actively consume.

The fresh tuna samples were caught by the few local longline fisherman and were purchased along Route 1 (the main road) near Tafuna. The sample pool consists of various species, including Skipjack Tuna (*Katsuwonus pelamis)*, Yellowfin Tuna (*Thannus albacares,* and Bigeye Tun*a* (*Thannus obesus*), all presented generally as tuna. Each of the samples reported is a result of the mean of 2 independently processed subsamples, and unless noted otherwise the results of the two samples were reasonably similar.

Figure 3.2: Typical 11.5 oz. Can of Corned Beef (Pisupo)

Heavy Metal Contaminant Analyses for Canned Tuna and Corned Beef

The product in each can was individually homogenized in a high-powered blender and two samples from each can were analyzed for 41 different elements by Quadrupole ICP-MS (ThermoFisher Scientific iCAP Q, with CCT option). For each sampled, approximately 0.2g of tissue was collected and digested in reverse aqua regia (1 part trace metal grade 12M hydrochloric acid and three parts trace metal grade 15.6 M nitric acid) on a 120°C hotplate for 24 hours. Following the digestion period, each sample was then diluted up to a volume of 5 mL with nitric acid. Mercury (Hg) concentration was measured separately based on US EPA method 1631E, with on-line Hg(0) cold vapor generation by $SnCl_2$ reduction. Maximum concentrations detected in fish and beef samples are reported in mg of metal contaminant per kilogram of wet tissue weight (e.g. parts per million or ppm). Analyses were conducted by the Keck Laboratory at Arizona State University.

Estimation of Consumption Risk

For each of the measured heavy metals with established oral reference doses (US-EPA IRIS database 2023), consumption risk was evaluated by calculating the associated Action Levels based on average Samoan adult body weight and weekly consumption rates of canned tuna and canned corned beef. Oral reference doses represent the maximum acute recommended human exposure and assumes static chronic consumption over time coupled with minimal change in body composition over time. To directly compare oral reference doses to measured metal concentrations, the oral reference doses are used to calculate Action Levels, which is a screening level threshold that corresponds to the maximum metal concentration in fish recommended to most likely avoid long-term negative impacts. Action levels were calculated using the same consumption rate for all consumables reported so that all the samples could be compared more equitably.

Action levels are calculated as follows:

Action Level (ppm) = (oral reference dose in mg kg-1 body weight x body weight (kg))/(serving size (kg) x # servings per week/7 days per week).

Survey of Canned Corned Beef (Pisupo) consumption rates

To calculate action levels more accurately, 37 adult individuals of Samoan decent were surveyed to assess their rates of pisupo consumption (Appendix 3.1: Survey). Of the 37 survey respondents, 33 people reported regular consumption of canned corned beef, ranging from ¼ of an 11.5 oz can to 2 - 11.5 oz cans per sitting, with a range of once to seven times per week. Several families reported consuming one 11.5 oz (~0.33 kg) can per sitting, which supports using a consumption rate of 0.3 kg three times per week for all samples, which is the same consumption rate utilized when calculating action levels for all fish samples. Using this consistent consumption rate allows a more direct comparison with canned tuna. IRB approved 6/13/18; STUDY00008374: American Samoa Seafood and Pisupo Consumption Survey.

Wilcoxin Sign Rank Test - Statistical Analyses Comparing Metal Concentrations Between Canned Tuna and Canned Corned Beef

I applied the Wilcoxin Sign Rank Test (non-parametric t-test on ranks) to assess the differences in heavy metal concentrations between canned tuna and canned corned beef samples. This test was chosen due to the non-parametric nature of the data, small sample sizes, and occasional unevenness of the number of samples. These statistics were calculated in R Studio (Version 2023.06.0+421).

Results

A total of 41 metal elements were analyzed. This study reports 13 of the most relevant metals, of which 11 have oral reference doses established by the Environmental Protection Agency (EPA): aluminum (Al), arsenic (As), cadmium (Cd), chromium (Cr), mercury (Hg), manganese (Mn), nickel (Ni), lead (Pb), selenium (Se), uranium (U), zinc (Zn). Of these 11 elements, mercury was the only element to exceed calculated action levels in two of the fourteen tuna cans that were tested, and none of the measured concentrations exceeded the action levels for any of the corned beef samples (Table 3.1).

Table 3.1

Heavy Metals: Maximum Concentrations and Action Levels in Canned Tuna and Corned Beef

		Maximum concentration (ppm)				
Metal		[c]n	Max	Average (Standard Deviation)	Action Level	p-value
Sodium (Na)						3.49 e^ -7
	Tuna	14	6191	2483 (1165)	NA	
	Corned Beef	14	8090	6553 (716)		
Aluminum (Al)						3.4 e^ -5
	Tuna	14	5.1	1.46 (1.4)	778	
	Corned Beef	14	1.1	0.5 (0.2)		
Arsenic (As)						NA
	Tuna	14	0.74	0.53 (0.12)	[b]0.23	
	Corned Beef	0	<LOD	<LOD		
Cadmium (Cd)						5.16 e^ -5
	Tuna	14	0.04	0.001 (0.006)	2.3	
	Corned Beef	6	0.0008	0.0005 (0.0002)		
Chromium (Cr)						0.02
	Tuna	14	0.57	0.07 (0.14)	2.3	
	Corned Beef	14	0.09	0.04 (0.02)		
ron (Fe)						4.99 e^ -8
	Tuna	14	10.1	7.9 (1.2)	NA	
	Corned Beef	14	25	18.4 (3.7)		
Mercury (Hg)						4.99 e^ -8
	Tuna	14	*0.12	0.06 (0.03)	*0.078	
	Corned Beef	1	0.001	0.001 (0)		
Manganese (Mn)						0.18
	Tuna	14	0.08	0.06 (0.02)	109	
	Corned Beef	14	0.09	0.05 (0.02)		
Nickle (Ni)						0.8
	Tuna	4	0.3	0.08 (0.15)	16	
	Corned Beef	1	0.06	0.06 (0)		
Lead (Pb)						0.09

	Tuna	14	0.78	0.34 (0.23)	3.9	
	Corned Beef	14	1.4	0.55 (0.34)		
Selenium (Se)						NA
	Tuna	13	0.27	0.22 (0.03)	3.9	
	Corned Beef	0	<LOD	<LOD		
Uranium (U)						2.79 e^ -5
	Tuna	14	0.001	0.0006 (0.0002)	2.3	
	Corned Beef	11	0.0003	0.0002 (0.0003)		
Zinc (Zn)						4.99 e^ -8
	Tuna	14	12	5 (2)	233	
	Corned Beef	14	31	26 (3)		

mg/ kg wet weight = ppm. <LOD = below level of detection.

NA = no oral reference dose available.

14 cans of canned tuna and 14 cans of canned corned beef were analyzed. The sample concentration is reported as the mean of the two subsamples from each can.

[c] n= the number of fish with measurable concentrations of the respective metal.

The maximum mercury (Hg) concentration for all groups exceeded the calculated action level based on a 100 kg adult show consumes 0.3 kg servings of fish muscle tissue 3 times per week.

NA = oral reference dose not available, therefore action level cannot be calculated.

[a] The EPA reports an oral reference does for inorganic arsenic, which is toxic and incomparable the less toxic organic form typically found in fish tissue.

[b] Action level is for more toxic inorganic form; fish contain organic As.

All Action Levels here are calculated from the EPA's Integrated Risk Information System (IRIS) oral reference dose database.

*The maximum concentration measured in canned tuna exceeded calculated action levels.

Heavy Metals in Canned Tuna vs. Canned Corned Beef

Measurable concentrations of metals were identified in most of the samples of both tuna and corned beef. The canned tuna samples contained all the metals reported in Table 3.1 (Na, Al, As, Cd, Cr, Fe, Hg, Mn, Ni, Pb, Se, U, Zn), and the canned corned beef samples contained all the same metals except for arsenic (As) and selenium (Se), which were below levels of detection.

Sodium (Na)

All samples contained measurable concentrations of sodium. The highest concentrations of sodium were found in the canned corned beef, with a maximum of 8090 ppm and a mean of 6553 (+/- 716). The cans of tuna had a maximum of 6191 ppm, with a mean of 2483 (+/- 1165), and the fresh tuna had a maximum of 1144 ppm, and a mean of 451 (+/- 229). The Wilcoxin sign rank test indicates significant differences in chromium concentrations between canned tuna and canned corned beef ($p = 3.49\ e^{\wedge -7}$). Fresh tuna had a maximum concentration of 1144 ppm, and a mean sodium concentration of 451 +/- 229 ppm. Renuka et al. 2017 reported mean sodium concentrations in Yellowfin Tuna of 677 +/- 0.92 ppm.

Table 3.2
Sodium (Na): Comparing Canned Corned Beef, Canned Tuna, and Fresh Tuna

Sample source	Max	Average (Standard Deviation)
Canned corned beef	8090	6553 (716)
Canned tuna	6191	2483 (1165)
[a]Fresh tuna	1144	451 (229)

mg/ kg wet weight = ppm
[a] Evaluated during this study

Aluminum (Al)

Aluminum was measured in all canned tuna and canned corned beef samples. In tuna, the mean aluminum concentration was 1.46 +/- 1.4 ppm and ranged from 0.56 ppm to 5.1 ppm. In beef, the mean aluminum concentration was 0.5 +/- 0.2, with a range of 0.33 ppm to 1.1 ppm. The Wilcoxin sign rank test indicates significant differences in chromium concentrations between canned tuna and canned corned beef (p = $3.4^{\wedge-5}$).

Arsenic (As)

Total arsenic was measured in all tuna samples but was below levels of detection for all the canned beef samples. The mean arsenic concentrations in tuna were 0.53 +/- 0.12 ppm, with a range of 0.32 ppm to 0.74 ppm.

Cadmium (Cd)

All 14 cans of tuna contained measurable concentrations of cadmium, with a range of 0.012 ppm to 0.036 ppm. Six of the 15 corned beef cans contained measurable concentrations, with a range of 0.0002 ppm to 0.0008 ppm. The Wilcoxin sign rank test indicates significant differences in chromium concentrations between canned tuna and canned corned beef
(p = $5.16^{\wedge-5}$).

Chromium (Cr)

Chromium was measured in all canned tuna and canned beef samples. In tuna, the mean chromium concentration was 0.07 +/- 0.14 ppm and ranged from 0.02 ppm to 0.57 ppm. In beef, the mean chromium concentration was 0.04 +/- 0.02, with a range of 0.02 ppm to 0.09 ppm. The Wilcoxin sign rank test indicates significant differences in chromium concentrations between canned tuna and canned corned beef (p = 0.02).

Iron (Fe)

Iron was measured in all canned tuna and canned beef samples. In tuna, the mean iron concentration was 7.88 +/- 1.2 ppm and ranged from 6.4 ppm to 10.1 ppm. In beef, the mean iron concentration was 18.2 +/- 3.7, with a range of 13.2 ppm to 25 ppm. The Wilcoxin sign rank test indicates significant differences in iron concentrations between canned tuna and canned corned beef (p = 4.99 $^{\wedge-8}$).

Mercury (Hg)

Mercury was measured in all 14 of the canned tuna samples. In canned tuna, the mean mercury concentration was 0.056 +/- 0.028 ppm and ranged from 0.025 ppm to 0.118 ppm. For the canned beef samples, one subsample of one sample resulted in 0.001 ppm, but the other subsample and all of the other samples were below the level of detection.

Manganese (Mn)

Manganese was measured in all canned tuna and canned beef samples. In tuna, the mean manganese concentration was 7.88 +/- 1.2 ppm and ranged from 6.4 ppm to 10.1 ppm. In beef, the mean manganese concentration was 0.06 +/- 0.02, with a range of 0.03 ppm to 0.08 ppm. The Wilcoxin sign rank test indicates no significant differences in manganese concentrations between canned tuna and canned corned beef corned (p = 0.18).

Nickel (Ni)

Four of the tuna samples had measurable concentrations of nickel. For three of the samples only one of the replicate subsamples had measurable concentrations of 0.005 ppm, 0.006 ppm, and 0.012 ppm. One sample appears to be an outlier with an approximate concentration of 0.3 ppm, where replicate subsample one had a concentration of 0.011 ppm and replicate subsample two had a concentration of 0.59 ppm. Only one replicate subsample of beef had measurable concentrations of nickel of 0.064 ppm.

Lead (Pb)

Lead was measured in all canned tuna and canned beef samples. In tuna, the mean lead concentration was 0.34 +/- 0.23 ppm and ranged from 0.008 ppm to 0.78 ppm. In beef, the mean lead concentration was 0.55 +/- 0.34, with a range of 0.18 ppm to 1.4 ppm. The Wilcoxin sign rank test indicates significant differences in lead concentrations between canned tuna and canned corned beef (p = 0.09)

Selenium (Se)

Selenium was measured in 13 of 14 tuna samples but was below levels of detection for all the canned beef samples. I In tuna, the mean selenium concentration was 0.22 +/- 0.03 ppm and ranged from 0.17 ppm to 0.27 ppm.

Uranium (U)

Uranium was measured in all canned tuna samples and 11 of the 14 canned beef samples. In tuna, and the mean uranium concentration was 0.0006 +/- 0.0002 ppm and ranged from 0.0004 ppm to 0.00095 ppm. For the beef samples, only one of the replicate subsamples had detectable levels, and the mean uranium concentration was 0.00015 +/- 0.00008 ppm, with a range of 0.00007 ppm to 0.0003 ppm. The Wilcoxin sign rank test indicates significant differences in uranium concentrations between canned tuna and canned corned beef ($p = 2.79\ ^{\wedge -5}$)

Zinc (Zn)

Zinc was measured in all canned tuna and canned beef samples. In tuna, the mean zinc concentration was 5.2 +/- 2.1 ppm and ranged from 3.3 ppm to 11.6 ppm. In beef, the mean zinc concentration was 26 +/- 3.5 ppm, with a range of 19.4 ppm to 30.7 ppm. The Wilcoxin sign rank test indicates significant differences in zinc concentrations between canned tuna and canned corned beef ($p = 4.99^{\wedge -8}$)

Mercury in canned tuna vs. fresh tuna

Two out of 14 cans of tuna had measured concentrations of total mercury that exceeded the calculated action levels of 0.078 ppm, with concentrations of 0.118 ppm and 0.091 ppm, respectively. The overall range of mercury concentrations in the cans of tuna was 0.025 ppm – 0.118 ppm, and additionally four cans had concentration that exceeded 0.07 ppm, which is close to, but does not exceed the calculated action level of 0.078 ppm (Table 3.3). Five out of 15 of the fresh tuna samples exceeded the calculated action level of 0.078 ppm, with concentrations of 0.117 ppm, 0.184 ppm, 0.24 ppm, 0.158 ppm, and 0.087 ppm (Table 3.4). Result of Kruskal-Wallis analyses indicates that there is no significant difference between mercury concentrations in fresh and canned tuna (W = 151, p = 0.1103).

Table 3.3
Mercury (Hg): Individual Canned Tuna Sample Concentrations

Canned Tuna Sample number	Hg Concentration (ppm)	Action Level
1	0.033	0.078
2	0.036	
3	0.063	
4	0.076	
5	0.048	
6	0.037	
7	*0.091	
8	*0.118	
9	0.038	
10	0.033	
11	0.072	
12	0.025	
13	0.074	
14	0.073	

mg/ kg wet weight = ppm
*Two canned tuna samples exceeded the action level
Concentrations given are the mean of two subsamples per sample

Table 3.4
Mercury (Hg): Individual Fresh Tuna Sample Concentrations

Fresh Tuna Individual by Species	Hg Concentration (ppm)	Action Level
1 - Skipjack Tuna	*0.117	0.078
2 - Skipjack Tuna	0.002	
3 - Skipjack Tuna	0.036	
4 - Skipjack Tuna	*0.184	
5 - Skipjack Tuna	0.041	
6 - Skipjack Tuna	0.044	
7 - Skipjack Tuna	*0.24	
8 - Skipjack Tuna	0.046	
9 - Yellowfin Tuna	0.06	
10 - Yellowfin Tuna	0.02	
11 - Yellowfin Tuna	*0.158	
12 - Yellowfin Tuna	0.021	
13 - Bigeye Tuna	0.056	
14 - Bigeye Tuna	0.024	
15 - unknown spp.	*0.087	

mg/ kg wet weight = ppm
*Five of the fresh tuna samples exceeded the action level.
Concentrations given are the mean of two subsamples per sample

Discussion and Future Directions

Most of the heavy metal concentrations measured in canned corned beef, canned tuna, and fresh tuna muscle tissue for this study are within ranges considered safe for human consumption based on calculated action levels for adult Samoans consuming 0.3 kg of fish or beef 3 times per week, except for mercury which exceeded action levels in several samples of canned tuna and fresh tuna.

Heavy Metals in Canned Corned Beef (pisupo) vs. Canned Tuna Mercury (Hg)

Mercury is the only heavy metal that had measured concentrations that exceeded calculated action levels. It is the most commonly studied contaminant in food, particularly in fish, due to its ubiquitous presence globally and its potential for toxicity.

The current study measured maximum mercury concentrations in canned tuna of 0.118 ppm, which is greater than the calculated action level for an adult Samoan of average weight (100 kg) consuming 0.3 kg of tuna three times per week, which is 0.078 ppm. Other studies reported a relatively wide range of mercury concentrations in canned tuna. Emami et al. 2005 reported a range 0.043 to 0.253 ppm. A single sample of canned tuna in oil resulted in a maximum of 0.04 ppm (Kowalksa et al. 2020). Canned tuna from Canada and India reported maximum concentrations of 0.6 ppm and 0.62 ppm, respectively. (Mahalakshmi et al. 2012). Canned tuna from Turkey found a range of below level of detection to 1.14 ppm (Mol 2011). Canned tuna from Italian market found a range of below level of detection to 0.21 ppm (Russo et al. 2013). Various brands of canned tuna purchased from Georgia and Alabama in the U.S. resulted in a concentration range of 0.053 ppm to 0.74 ppm (Ikem and Egiebor 2005). The highest reported concentration was 1.14 ppm in canned tuna from Turkey.

The current study measured mercury maximum concentrations in canned corned beef of 0.001 ppm, which was identified in only one subsample of a sample. Kowalksa et al. 2020 measured a range from 0.00003 ppm – 0.00007 ppm. Defouli et al. 2018 measured mercury in 10 different unnamed brands of canned tuna with origins in the Persian Gulf, and an overall range of 0.076 ppm to 0.31 ppm. Morshdy et al. (2023) identified ranges from below level of detection to 0.11 ppm, and three brands of canned corned beef had an overall range of concentrations from below detection levels to 0.005 ppm (Abinotami et al. 2023). The overall range of mercury concentrations in canned corned beef from the reviewed studies is from below level of detection to the greatest measured concentration of 0.31 ppm.

Fish are exposed to mercury from industrial pollution emission into waterways (Bełdowska 2016), and consistently harbor greater mercury concentrations than beef sources. The results of this study support this trend with maximum concentrations in canned tuna of 0.12 ppm, and in canned corned beef of 0.001 ppm.

Sodium (Na)

While sodium is an essential nutrient and is not necessarily considered toxic in the same sense as some of the other metals that we evaluated, excessive sodium (chloride) in one's diet can contribute to negative health outcomes, such as high blood pressure, heart disease, stroke, fatigue, and in excessive amounts can cause death. Sodium does not occur in its free metallic state in the environment due to its high reactivity, but when bound with chloride it becomes and essential mineral for the human body in small amounts. The ICP-MS detects sodium ions (Na^+), which constitutes approximately 40% - 50% of the sodium chloride compound (American Chemistry Society 2023, Harvard School of Public Health 2023, Libretexts Chemistry 2023) we evaluated sodium levels and compared to the nutritional data reported on the cans.

Sodium levels reported on the can labels for both canned tuna and canned corned beef fell within the minimum and maximum range measured in the laboratory for this study. When converted to mg/ kg (ppm), the nutritional information on the tuna cans report 3160 ppm of sodium per serving; this study measured a mean of 2483 ppm and ranged from 1272 ppm to 6191 ppm, resulting in the mean being slightly higher that what is reported on the can. When converted to mg/ kg (ppm), the nutritional information on the cans of corned beef reports 5360 ppm of sodium per serving; this study measured a mean of 6553 +/- 716 ppm and ranged from 5153 ppm to 8090 ppm (Table 3.2), resulting in the mean being slightly higher than what is reported on the can. The FDA recommends that the average adult American consumes less than 2,300 mg per day (based on the ionic sodium reported on nutrition labels) (FDA – Sodium in your diet 2023). Processed canned foods often have high levels of sodium unless specifically reported otherwise. Salt helps in the preservation of food and enhances flavor and palatability. But also, can contribute to negative health outcomes, therefore when considering health outcomes, it can be helpful to understand that there is some variability from what nutrition labels might be reporting.

There are few publications that have reported heavy metal concentrations in canned tuna, and even fewer that have reported them in canned corned beef.

Aluminum (Al)

Aluminum is the most abundant metal in Earth's crust. It is an unstable element, readily binding to other elements such as oxygen, silicon, and fluorine. It is used for many human applications, including in medications, cosmetics, and food items, in items such as pots and pans, building supplies, and in explosive and fireworks (CDC- ATSDR - Aluminum Toxicity 2023) When ingested in elevated concentrations, aluminum can lead to kidney and liver damage, and can contribute to reduced bone mineralization (Rahimzadeh et al. 2022).

The current study measured maximum aluminum concentrations in canned tuna of 5.1 ppm. This was higher than

canned tuna that was sourced from Canada and India, which found maximum concentrations of 1.8 ppm and 3.7, respectively (Mahalakshmi et al. 2012). The current study measured maximum concentrations of aluminum in canned corned beef of 1.1 ppm, which is in the range of concentrations that Morshdy et al. 2023 found in canned corned beef sourced from stores in Egypt, which was from 0.15 ppm to 7.95 ppm (Morshdy et al. 2023). Most of the reviewed studies did not report aluminum concentrations in their results, and overall, the results between canned tuna and canned corned beef overlapped.

Arsenic (As)

Arsenic is widely distributed element, occurring in both the inorganic and organic forms, which result in drastically different toxicity outcomes. Inorganic arsenic is found in sediments and in water and can be highly toxic and carcinogenic (Chung et al. 2014), whereas organic arsenic is mostly found in fish and shellfish and is considered to result in low toxicity (CDC- ATSDR Arsenic Factsheet 2023). The US – EPA/ FDA have created an oral reference dose for aluminum phosphate, which is a highly toxic form of inorganic arsenic used for pest control (US – EPA/ FDA Arsenic 2023), and thus is not directly comparable to fish sample concentrations.

The current study measured maximum arsenic concentrations in canned tuna of 0.74 ppm, which is higher than was found in canned tuna from Iran, which had a range of concentrations from 0.05 to 0.21 ppm (Emami et al. 2005). A single sample of canned tuna in oil resulted in a maximum of 0.77 ppm (Kowalksa et al. 2020), which is close to what we measured in this study. Various brands of canned tuna purchased from Georgia and Alabama in the U.S. resulted in a concentration range of 0 ppm to 1.72 ppm (Ikem and Egiebor 2005), which is a range that also encompasses the results of this study.

For the current study all of the arsenic concentrations in canned corned beef were lower than the level of detection. Kowalksa et al. 2020 also found low concentrations of arsenic, ranging from 0.002 ppm – 0.003 ppm. Overall arsenic concentrations are low in canned corned beef, and much higher in canned tuna due to their exposure in (sea)water, by which they uptake arsenic by ingesting water through their mouth and gills (Kumari et al. 2017).

Cadmium (Cd)

Cadmium was one of the most frequently reported metals in the studies reviewed. It enters the environment largely from mining, and less so from fires and volcanos, and is distributed by wind and water movement. In elevated concentrations, cadmium can cause digestive distress, over time can build up in the kidneys causing kidney disease and fragile bones, and can cause cancer (CDC- ATSDR Cadmium Factsheet 2023). While fish consumption adds to overall cadmium consumption, the main vectors of human exposure are from ingesting wheat, rice, and potatoes, and from smoking cigarettes ((CDC- ATSDR Cadmium Factsheet 2023); Djedjibegovic et al. 2020).

The current study measured maximum concentrations of cadmium in canned tuna of 0.036 ppm, which was higher than Mahalakshmi et al. 2012 measured in canned tuna from Canada and India, which reported maximum concentrations of 0.02 ppm and 0.025, respectively. Emami et al. (2005) measured a range that encompassed both of our studies, with a range of 0.005 – 0.07 ppm. A single sample of canned tuna in oil resulted in a maximum of 0.01 ppm (Kowalksa et al. 2020), which is in the range of concentrations identified in the other studies reported here. Canned tuna samples from Turkey found a range of below level of detection to 0.09 ppm (Mol 2011), and canned tuna from Italian markets found a range of below level of detection to 0.07 ppm (Russo et al. 2013). Various brands of canned tuna purchased from Georgia and Alabama in the U.S. resulted in a concentration range of 0 ppm to 0.05 ppm (Ikem and Egiebor 2005). The overall range of cadmium identified in the above studies is from undetectable to 0.09 ppm.

The current study measured maximum concentrations in canned corned beef of 0.0008 ppm. Nasser (2014) measured cadmium concentrations from 0.16 ppm - 0.61 ppm, and Kowalksa et al. 2020 measured concentrations from 0.009 ppm – 0.02 ppm, both higher than was measured in the current study. Canned corned beef from Egypt had a range from below level of detection to 0.09 ppm (Khalafalla et al. 2016), and three brands of canned corned beef had an overall range of concentrations from 0.001 ppm to 0.01 ppm (Abinotami et al. 2023). The overall range of cadmium in canned corned beef identified in the above studies is from 0.0008 ppm to 0.09 ppm. The maximum concentrations of cadmium in both canned tuna and canned corned beef in the above-mentioned studies were 0.09 ppm.

Chromium (Cr)

Chromium is a widely distributed element, with human sources resulting from mining and industrial combustion and in certain forms, such as hexavalent chromium, can be highly toxic, resulting in stomach cancers in humans (CDC- ATSDR Chromium 2023). Most other forms of chromium are not as acutely toxic, but accumulation overtime might negatively impact epithelium in the stomach (Aslam, S and AM Yousafzai 2017).

The current study measured maximum concentrations of chromium in canned tuna of 0.57 ppm, which is lower than what was measured in a single sample of canned tuna in oil which had a maximum of 0.99 ppm (Kowalksa et al. 2020). Various brands of canned tuna purchased from Georgia and Alabama in the U.S. resulted in a concentration range of 0 ppm to 0.07 ppm (Ikem and Egiebor 2005), lower than was reported for the other two mentioned studies.

The current study measured maximum concentrations in canned corned beef of 0.09 ppm, which was less than the ranges that Kowalksa et al. (2020) measured, which was 0.2 ppm to 0.32 ppm. Overall, corned beef had higher concentrations of chromium than fish, which is consistent with established results (NIH Chromium 2023).

Iron (Fe)

Iron enters the environment primarily through weathered rocks and sediments. Iron is an essential element for the human body (Abbaspour et al. 2014), and iron deficiency is common in the United States, particularly for menstruating women. But iron absorption is complex; excess iron is recycled throughout the body and is stored primarily in the liver, spleen, and bone marrow and in excess can lead to digestive and cardiovascular issues and increased risk of cancer (Abbaspour et al. 2014). Humans absorb approximately 5% to 35% of the iron that is consumed, further complicating overall iron needs and subsequent toxicity. The National Institutes of Health (2023) recommend that adult men consume 8 mg per day and that women consume 18 mg per day but take caution specifically when taking iron supplements to avoid potential toxicity, which can result in severe gastrointestinal stress and liver damage.

The current study measured a range of iron concentrations in canned tuna of 6.7 to 10.1 ppm. Canned tuna from Turkey was was below levels of detection to 80.7 ppm (Mol 2011). Various brands of canned tuna purchased from Georgia and Alabama in the U.S. resulted in a concentration range of 0.01 ppm to 88.4 ppm (Ikem and Egiebor 2005). The results of the current study falls within the range of both the cans from Turkey and the U.S., but has less variation.

The current study measured maximum iron concentrations in canned corned beef of 25 ppm, which is in the range of canned corned beef purchased from Brazil in Saudi Arabia, with a of range 11.8 ppm - 39.1 ppm (Nasser 2014). Three brands of canned corned beef imported from Brazil to Nigeria had an overall range of concentrations from 9.5 ppm to 14.3 ppm (Abinotami et al. 2023), lower than the maximum concentrations measured in the current study. Overall, the highest concentrations in tuna for these studies was 88.4 ppm, and 39.1 ppm for corned beef. Cleveland Clinic (2023) confirms that meat products are the highest food sources contributing to iron intake.

Manganese (Mn)

Manganese occurs naturally in many rocks and sediments bound to other substances such as oxygen, sulfur, and chlorine. For industrial purposes, manganese is used steel production to improve its strength, and is used in other manufacturing processes, including fireworks, fertilizers, paints, and cosmetics (CDC- ATSDR – Toxilogical Profile for Manganese 2023b). It is an essential element in limited amounts, but in elevated concentrations can lead to neurological disorders (CDC- ATSDR Manganese 2023a. Humans are primarily exposed by consuming beans, nuts, grains, and tea, and from nutritional supplementation (CDC- ATSDR – Toxilogical Profile for Manganese 2023b).

The current study measured maximum manganese concentrations in canned tuna of 0.08 ppm, less than a single sample of canned tuna in oil with a maximum of 0.26 ppm (Kowalksa et al. 2020). Various brands of canned tuna purchased from Georgia and Alabama in the U.S. resulted in a concentration range of 0.08 ppm to 0.63 ppm (Ikem and Egiebor 2005).

The current study measured maximum manganese concentrations in canned corned beef of 0.09 ppm, less than measurement from Kowalksa et al. (2020), which ranged from 0.16 ppm to 0.28 ppm. Similar concentrations were measured in canned tuna and canned corned beef samples, and among the most common food sources of manganese is from grains, shellfish, nuts, legumes, spices, coffee and tea (CDC- ATSDR Manganese 2023).

Nickel (Ni)

Nickel is found in all types of soil in the environment, typically bound to oxygen or sulfur, and is emitted by volcanic activity. Humans use nickel in various applications, including nickel plating, battery making, and ceramic coloring. It is also a byproduct of emissions from trash incinerators and powerplants. Nickel tends to bind strongly to sediments, reducing its overall bioavailability. Foods, including chocolate, soy, legumes, nuts and oatmeal are among the major sources of nickel in humans (CDC- ATSDR Public Health Statement for Nickel 2005; Sharma 2013). Nickel is the greatest metal food allergen in humans most commonly resulting in dermatitis from contact, and resulting in kidney, liver, immune, and reproductive harm when ingested in excess (CDC- ATSDR Tox Facts for Nickel 2023; Sharma 2013).

The current study measured maximum of nickel concentrations in canned tuna of 0.3 ppm (with the second highest concentration being 0.01 ppm). Kowalksa et al. 2020 found a single sample of canned tuna in oil to contain a maximum of 0.1 ppm. Various brands of canned tuna purchased from Georgia and Alabama in the U.S. resulted in a concentration range of 0 ppm to 0.12 ppm (Ikem and Egiebor 2005). The maximum concentration from this study was higher than the other reviewed studies, but the second highest was in the range of the other studies.

The current study measured maximum concentrations of nickel in canned corned beef of 0.064 ppm maximum. Kowalksa et al. 2020 found that canned corned beef ranged from 0.002 ppm – 0.004 ppm, which is lower than the current study.

Lead (Pb)

Lead is found in limited amounts naturally in rocks and sediments, but due to human activity such as industrial combustion and intentional use, lead is among the most currently toxic metal to humans. Humans have a long history of using lead in manufacturing of household products and cosmetics, creating opportunities for greater contact with this metal (US-EPA Learn about Lead 2023; Zhang et al. 2011). In elevated concentrations, lead can lead to brain and nervous system damage, cognitive and speech challenges, and (CDC- ATSDR Health Effects of Lead Exposure (2022). Children are particularly suseptible to lead poising as they are growing (CDC- ATSDR Lead Poisoning Prevention 2022).

The current study measured maximum of lead concentrations in canned tuna of 0.78 ppm, which is higher than Kowalksa et al. 2020 reported of a single sample of canned tuna in oil resulted in a maximum of 0.37 ppm. Emami et al. 2005 reported a much lower range of 0.016 – 0.073 ppm. Canned tuna from Canada and India; Maximum concentrations from Canada was 0.01 ppm, and 0.09 ppm in India. (Mahalakshmi et al. 2012). Canned tuna from Turkey found a range of below level of detection to 4.13 ppm (Mol 2011). The units were double checked because this result was so much higher than any of the other results presented here. Canned tuna from an Italian market found a range of below level of detection to 0.51 ppm (Russo et al. 2013). Various brands of canned tuna purchased from Georgia and Alabama in the U.S. resulted in a concentration range of 0 ppm to 0.03 ppm (Ikem and Egiebor 2005). There was a wide range of lead concentrations in tuna, which ranged from below levels of detection to 4.13 ppm found in samples from Turkey.

The current study measured maximum concentrations of lead in canned corned beef of 1.4 ppm, which is higher than what Nasser (2014) measured, which was a range of 0.36 ppm - 1.12 ppm, and also higher than Kowalksa et al. (2020), which ranged from 0.03 ppm – 0.43 ppm. Canned corned beef from Egypt had a range from below level of detection to 0.63 ppm (Khalafalla et al. 2016). Three brands of canned corned beef had an overall range of concentrations from 1.7 ppm to 2.5 ppm (Abinotami et al. 2023). Based on these studies, the range of lead in canned corned beef ranges from 0.03 ppm to 2.5 ppm. Meat consumption is not a major source of lead in food, direct deposition to plant crops are much more likely to contribute to elevated lead levels, which is also likely why cows exhibited higher lead concentrations in this study (CDC Lead in Food, Cosmetics, and Medicine 2023).

Selenium (Se)

Coal mining is the source of most of the environmental selenium, and in elevated concentrations can result in deformities in growing offspring and overall reproductive damage (Lemly 2008). Among the most common food sources of selenium are from Pork, beef, turkey, chicken, fish, shellfish, eggs, nuts, and legumes (National Institution of Health (NIH) Selenium 2021).

The current study measured maximum of selenium concentrations in canned tuna of 0.27 ppm, and maximum concentrations in canned corned beef of selenium below level of detection for all samples. None of the other studies reviewed reported selenium concentrations. The NIH (2023) reports that sources of selenium are more likely from beef (vs fish) sources, but that is not the trend that was observed for the current study.

Uranium (U)

Uranium is a naturally occurring radioactive element that occurs in small amounts in the environment but is liberated into the ecosystem from mining practices, where it persists (CDC- ATSDR Uranium Toxicity (2012). Uranium toxicity is highest for people who are exposed to uranium mining, milling, and fabrication. The most common outcomes of respiratory inhalation are respiratory disease, and the most common outcomes from ingestion is renal failure and potential death (CDC- ATSDR What Are the Physiological Effects of Uranium Exposure? 2023).

The current study measured maximum of uranium concentrations in canned tuna of 0.00095 ppm maximum and measured maximum concentrations of uranium in canned corned beef of 0.0003 ppm. Uranium was undetectable in both canned tuna in oil and in canned corned beef samples (Kowalksa et al. 2020). The other studies reviewed did not report uranium concentrations.

Zinc (Zn)

Zinc enters the environment naturally through geological weathering and is also released during smelting and mining activites. Zinc resists corrosion, therefore, has been used in manufacturing many commonly used products, including in propellants, fire retardants, fungicides, paints, batteries, and deodorants (Environmental Pollution Centers 2023; Zhang et al. 2011). It is an essential element for the human body, and among the most food sources are from beef, pork, dairy, fish, shellfish, legumes, and through fortification of infant products, but in elevated concentrations can result in urinary problems, neurological problems, and organ damage (CDC- ATSDR Zinc 2023).

The current study measured maximum of zinc concentrations in canned tuna of 11.6 ppm, which is below the maximum, but within the range of what Nasser (2014) measured, which was 8.3 ppm - 28 ppm (Nasser 2014). Canned tuna from Turkey found a range of 3.7 to 30 ppm (Mol 2011). Various brands of canned tuna purchased from Georgia and Alabama in the U.S. resulted in a concentration range of 0.14 ppm to 9.87 ppm (Ikem and Egiebor 2005). The overall range of zinc in canned tuna was 0.14 ppm to 30 ppm.

The current study measured maximum concentrations of zinc in canned corned beef of 30.7 ppm, which is higher than what Abinotami et al. (2023) measured in three brands of canned corned beef which had an overall range of concentrations from 17 ppm to 23 ppm. The resulting zinc concentrations overlapped between the canned tuna and canned corned beef samples, which supports previous research that indicates both beef and seafood are among the richest sources of zinc in human diets (NIH Zinc 2023)

Mercury in canned tuna vs fresh tuna

The *maximum* concentrations for both fresh and canned tuna for this study exceeded calculated action levels, but the *means* for both fresh and canned tuna fell below the action levels. The United States EPA - FDA's monitoring database of mercury levels in commercial fish and shellfish (EPA - FDA 1990-2012) informs public advice regarding the potential hazards from consuming various types of marine fish available for consumption in the United States, including for fresh and canned tuna. They currently report a maximum concentration of 1.8 ppm Hg for fresh tuna and a maximum of 0.89 ppm for canned tuna, and mean concentration of 0.39 ppm for fresh tuna and a mean of

0.13 ppm for canned tuna, which are both greater than maximum and means identified in this study and both exceed the action level calculated for the current study (Table 3.5). Overall, the results of the current study indicate lower maximum and mean concentrations of mercury than the US - EPA/ FDA monitoring database, measuring maximum mercury concentrations in canned tuna of 0.12 ppm and 0.12 ppm in fresh tuna (US – EPA/ FDA Mercury in Fish Monitoring Database 2023), compared to . This study measured the same maximum concentrations of 0.12 ppm in the Starkist canned tuna in oil samples, as well as 0.12 ppm in the collective fresh tuna samples. These results are not surprising given that the fish that the Starkist tuna plant are processing and selling are among the same stocks that are being sold and consumed locally.

Fresh tuna was analyzed raw, and canned tuna is cooked in the can. Considering that cooking meat might change mercury composition and overall metal concentrations, I researched the potential variation so we can make more informed comparisons. Ouedraogo and Amyot (2011) compared total mercury between raw, boiled, and fried tuna and found that raw tuna contained 1.37 +/- 0.62 ppm (dry weight), lower than tuna that had been boiled (1.57 +/- 0.76 ppm), and slightly higher than fried tuna (1.3 +/- 0.53). Chera-Anghel et al. 2023 reported concentrations less than 0.5 ppm for both raw and cooked tuna. Due to the similarities in mercury concentrations between raw and cooked tuna in previous studies, I assume direct comparability in this regard between fresh and canned tuna for this study.

Table 3.5
Mercury (Hg): Comparing Current Study Results to the US- EPA - FDA's Monitoring Database of Mercury Levels in Commercial Fish and Shellfish (EPA - FDA 1990-2012), Results for Canned and Fresh Tuna.

Sample source	Max	Average	Action Level
Canned Tuna[a] (EPA/ FDA)	*0.89	*0.13	0.078
Canned Tuna (Current study)	*0.12	0.06	
Fresh Tuna[b] (EPA/ FDA)	*1.8	*0.39	
Fresh Tuna (Current study)	*0.12	0.03	

* Measurements exceed calculated action levels for an adult Samoan consuming 0.3kg fish 3 times per week.
[a] Identified as Tuna (Canned, Light)
[b] Identified as Tuna (Fresh/ Frozen, All)

Kowalski (2020) notes that fish and fish products are among the most commonly tested food items due to their propensity for harboring toxic metals, particularly mercury, but there are few studies that investigate the comparisons between meat and fish products, and even fewer that have targeted specific products that the majority of a population are consuming.

This study is the first to investigate and compare heavy metal concentrations in commonly consumed canned corned beef, canned tuna, and fresh tuna in American Samoa. Based on the calculated action levels, an average weight (100 kg) adult individual of Samoan decent has minimal risk of heavy metal toxicity when considering single contaminant toxicity, with the potential exception of elevated concentrations of mercury in canned and fresh tuna. While only two of the cans of tuna exceeded action levels, four cans had measured concentrations above 0.07 ppm, and all the tuna cans had measurable concentrations of all the metals that were analyzed.

Canned Corned Beef (pisupo)

Most survey respondents reported that they love eating pisupo, including on respondent reporting that “pisupo is life”. Personal experience illustrated the prominence of pisupo in the American Samoan diet; at every celebration I attended, cans or cases of pisupo were given as gifts. It was frequent to see a person eating it straight from the can. When attending events, there was typically a blend of traditional Samoan cuisine combined with novel processed food items, and almost always, pisupo.

In many ways canned foods are an anomaly in American Samoa. Technically it is less labor intensive to acquire prepackaged foods than to forage or fish for fresh food, yet this is not the necessarily the driving force behind making particular food choices. It is common for fishermen to trade their labor-intensive catches for pisupo because a person having the resources to purchase packaged food reflects a sign of affluence in American Samoa (Simes 2018). While it is generally accepted that excessively consuming pisupo contributes to negative health outcomes, few people feel compelled to quit eating it. Only one family from our consumption survey reported that they no longer consume pisupo or have it in their house, reporting “because it is unhealthy, contains uric acids, has a lot of preservatives, fats, oils. It's red meat. Cow production has a large carbon footprint, it's bad for the environment.”

I considered the potential of migration of metals from the cans to the meat product but did not specifically focus on this. It is possible that metals might contaminate the meat inside, dependent on the type of can and its lining, and the conditions that the can endures, including temperature, humidity, age, and stress (Buculei et al. 2013). There is also evidence that canning and storing might affect the contents, including the alteration macronutrient profiles. For example, (Aberoumand 2023) measured nutrient and heavy metals concentrations and found that protein was greatest for fresh tuna and decreased over the course of 11 months from a mean of 22.66 % protein to a mean of 18.13 % protein, respectively, but didn't report dramatic changes in metal concentrations. Sulistiawaty et al. (2019) measured Iron and Zinc in tin can packaging in an acetic acid environment and a lipid rich environment and found that when exposed to oxygen, the cans leached these metals.

Further related to the can itself, I recommend that future studies evaluate potential organic contaminants that might migrate from the can lining to evaluate potential risk related to exposure to individual pollutants or a cocktail of pollutants, for example, potential endocrine disruptors such as BPA (Cunha et al. 2020, Siddique et al. 2021). Additionally, further researching organic pollutants in commonly consumed food items will help define the changes in concentrations over time and might help to clarify why studies are finding more organic pollutants when conducting laboratory analyses on environmental and fish samples.

Canned corned beef, canned tuna, and fresh tuna are commonly consumed food items for people in American Samoa, with survey respondents from the current study indicating that they consume fresh tuna at the same rate that they consume canned corned beef, 89%. This delicacy has only been a part of the Samoan diet for approximately 100 years. While it's generally accepted in American Samoa that as a highly processed food item, canned corned beef contributes to the islands’ top health issues, only one of our survey respondents reported that they quit eating pisupo due to concern of potential negative health outcomes.

CHAPTER 4

OVERALL DISCUSSION

Heavy metals and persistent organic pollutants impact coastal ecosystems and fish populations worldwide due to their ability to be transported long distances and to persist in the environment. All of the available published data regarding heavy metal concentrations in fish collected from American Samoa indicate that mercury (Hg) is the heavy metal of most concerns for people eating local fish, being the only metal to exceed calculated action levels, but the concentrations of mercury do not appear to be dramatically increasing or decreasing at this time. The source of the mercury in the fish is unclear; much of the global mercury deposition is due to coal combustion, which is still a major energy source globally and can be transported long distances atmospherically. Early studies measuring heavy metals in fish in American Samoa indicate that (total) arsenic (As) and lead (Pb) were potential contaminants of concern, but more advanced analyses revealed that the organic form of arsenic found in fish tissue is only approximately 0.5% of total arsenic, resulting in overall low toxicity risk. Although it is unclear why, the highest lead concentrations from all of the relevant studies were measured in 1990, but decreased dramatically in future studies, and have remained low (Government of American Samoa 1991, AS-EPA 1994, Peshut and Brooks 2005, US-EPA 2006, Morrison et al. 2015).

Few persistent organic pollutants were identified in fish tissue from any of the historical data, with the exception of specific polychlorinated biphenyl mixtures. Since many of the pollutants identified in this study have been banned since the 1970s, it might be hypothesized that there would be fewer persistent organic pollutants (POPs) in fish tissue over time, but the current study identified many more POPs than had been previously reported from American Samoa, including multiple types of polychlorinated biphenyls (PCBs), polychlorinated aromatic hydrocarbons (PAHs), pesticides, and phthalates in fish from all 12 taxonomic groups. It is possible that some of these pollutants are still entering the environment, or they might be moving around. A point of uncertainty is why prior to the current study the only persistent organic pollutants to be identified in fish were PCB mixtures (and DDD and DDE from one individual fish collected in 1990); there is evidence that these pollutants were measurable with the analytical techniques at the time, including evidence from fish caught near Australia and New Zealand but were not identified in the historical samples from American Samoa (Kannan et al. 1994, Lana et al. 2014).

This study started out by conducting analyses of heavy metals and persistent organic pollutants in locally caught fish, and to investigate potential risks associated with local fish consumption, yet upon interacting with the community I hypothesized that most of the Samoans were consuming as much if not more portions of canned corned beef daily than they were of fresh fish (survey results indicate that 89% of my respondents report consuming fish and 89% report consuming canned beef), therefore the analyses was expanded to include this food item. Additionally, these food choices are likely contributing directly to negative health outcomes including obesity and heart disease in the American Samoa population (Neuendorf et al. 2021).

According to the World Health Organization, Samoans are among the most obese populations on the planet; Ichiho et al. 2013 estimated almost a decade ago that ~93.5% of American Samoan adults are overweight or obese, and ~47.3% have diabetes.

Contaminants may interact with fat tissue in the body, and because of the lipophilic nature of organic pollutants, they tend to sequester in adipose tissue. While the individual is gaining weight, they are somewhat protected from the chemical effects, but can liberated into the blood stream upon fat loss (Lee 2017). The relationship between obesity and contaminant body burdens is complex and depends on the chemical in question. Lipophilic compounds accumulate in adipose tissue, and subsequently can be released into the blood stream over time, or rapidly due to weight loss or breastfeeding (Hue et al. 2006, La Merrill et al. 2013). Organic pollutants have been reported to be obesogenic and might impact the progression of diabetes (Jackson et al. 2018). According to the Institute for Health Metrics and Evaluation (IHME 2017) the top 3 risk factors driving death and disability combined in American Samoa are high-body mass index, high fasting plasma glucose, and dietary risks. These risk factors lead to the top two causes of death, heart disease and diabetes. The #1 cause of disability is diabetes. There is evidence that chronic background level exposure to persistent organic pollutants might contribute to insulin resistance, heart disease, and 9 diabetes later in life (Lee et al.2011). This relationship has been seen for among the Inuit communities in the Canadian Arctic and requires further investigation (Sighn 2018)

Obesity and chemical burdens are a complex issue; larger humans can theoretically (safely) intake a greater amount of chemicals than smaller humans, but if a person loses body fat, chemicals stored in fat cells can be liberated into the blood, potentially making it more bioavailable and toxic (Cheikh Rouhou et al. 2016). I investigated relationships between local pollution in American Samoa and the relatively recent shift in the people's diet, and while there are clearly relationships between the potential toxicity of consuming fish and how the perception of that affects a person's choice on what they decided to consume, the relationships look more like a web than individual variables that can be directly assessed. The story of local pollution, pollution in food, and the change in diet (and health) in American Samoa is a complex one influenced by trade and increases in available resources and subsequent waste, local industry, and relations with the US and other nations. Additionally, due to global reduction in overall tuna consumption and more specifically canned tuna consumption, the Starkist tuna plant in American Samoa has an uncertain future, which could potentially lead to another shift in dietary practices and would certainly be a shift economically as it is the processing facility provides the greatest number of jobs for the inhabitants of American Samoa and contributes greatly to the overall economy.

The results of this study indicate that the ingestion of chemical pollutants from fish and canned beef is not likely a primary driver for current prominent negative health outcomes, including obesity and type-two diabetes in American Samoa. Yet because the population experiences high rates of metabolic diseases, they might also be susceptible to elevated levels of toxic chemicals. As processed canned foods continue to be a prominent part of the Samoan diet, combined with increasing obesity and heart disease, it is important to maintain a current understanding of potentially toxic chemicals in the food items that we consume and how that might impact health outcomes over time.

Sources of Uncertainty and Future Directions

There are multiple sources of uncertainty to consider for this project. Starting with sample collection, I suggest that future studies specifically target resident fish both within and outside of Pago Pago harbor due to the history of pollution in the Harbor; this data will more accurately reflect the collection methods of the historical studies. It is unlikely that any fish collected for the current study were caught within the harbor, as it is discouraged to consume these fish, and these fish are intentionally caught for consumption. Additionally, pollutants are often transported atmospherically over long distances, resulting in measurable concentrations within fish tissue that might have originated from great distances; this trend is known for mercury, which is largely a result of burning coal in heavily industrialized countries.

There are many factors that might alter the results of risk calculations. This study assumes average body weight (100kg) and average consumption (0.3kg of the food item 3 times per week), therefore people who weigh more or less than the average will experience differences in risk. Children and reproductive women might have elevated risk as well, as some pollutants are known to negatively impact growing humans. Further, it can be challenging to directly compare the historical studies with each other and with the current study due to potential differences in analytical methods.

Due to its isolation, there are relatively few longitudinal studies that encompass the complexities of contaminants in food and food related practices in American Samoa, therefore, to preserve and document ongoing changes, I recommend that semi-regular monitoring of heavy metals and persistent organic pollutants in fish and other commonly consumed food items occur in the future. Additionally, I recommend that a follow up study be conducted to assess the populations of the pipi and tunagi clams, as the evidence that I gathered thus far indicates potential functional extinction of both populations, or perhaps just a dramatic shift in the consumption and sale of these clams in a very short period of time, or both. The bottom line, they used to be sold in abundance, and now they are not, and the locals (including previous clammers) don't seem to have an explanation.

This study assumes that all fish were the same size and age, ignoring that these metrics might contribute to the observed variability. There are several ways in which the laboratory analytics and subsequent action level calculations might not accurately reflect true risk. This dissertation does not correct for contaminant recovery rates, therefore underestimating contaminant concentrations. It is also important to note that many contaminants detected do not have established oral reference doses, and there are also likely contaminants present in fishes that were not able to be detected with GCMS, such as pharmaceuticals, or are present in levels below method or instrumentation detection limits.

The results of the current study exhibit a greater number of persistent organic pollutants in fish tissue than was identified in earlier studies, but it is unclear which proportion of that is due to an increasing number of these contaminants in the environment or due to advances in technology which result in greater detection pollutants. To properly assess ongoing trends in fish and novel food consumption and current risk, I suggest to periodically investigate local contaminant levels, in conjunction with monitoring rates. There was minimal information regarding the laboratory methods utilized to measure heavy metal and persistent organic pollutant concentrations. Different methods can result in variation in results, therefore it is recommended that future studies include methods to enhance the robustness of comparisons being made.

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APPENDIX A

SUPPORTING INFORMATION FOR CHAPTER 2

Supplementary Table 1:
Select Persistent Organic Pollutants: Maximum concentrations and action levels in fish muscle tissue

	Groups based on trophic -level dietary preferences				
	A Primarily herbivorous	B Plankton and small inverts.	C Macrobenthic organisms and small fish	D Pelagic organisms	Action Level
POP Category POP Compound					
PAHs					
anthracene	0.05	0.06	0.07	0.06	233
1-methylnapthalene	0.06	0.07	0.12	0.02	16
PCBs					
biphenyl	0.02	0.02	0.03	0.002	389
Pesticides					
carbaryl	0.01	0.005	0.09	0.002	78
DDD	0.01	0.0009	0.009	0.002	NA
DDT	NA	0.0006	NA	0.1	0.39
endosulphan (alpha isomer)	0.24	NA	0.13	0.67	6.7
hexachlorobenzene	0.003	NA	0.03	0.003	0.62
methoxychlor	0.93	0.38	0.33	0.67	3.9
mirex	0.008	0.009	0.04	0.004	0.16
trans- nonachlor (nonachlor/ chlorodane)	0.002	*1.3	0.001	0.0001	[a]0.39
Pthalates					

dibutyl phthalate	0.45	0.44	0.43	0.51	78
diethyl phthalate	0.02	0.01	0.02	0.005	622
di (2-ethylhexyl) phthalate	2.2	2.1	2.7	0.26	15.6

Compound concentrations are in mg/ kg (wet weight) = ppm

*One individual squirrelfish exceed [a]action level for trans- nonachlor

Supplementary Table 2:

Persistent Organic Pollutants (POP): Summary Table of Kruskal – Wallis rank sum test

POP Category	[a]KW Rank Sum Test
PAHs	0.67
PCBs	0.14
Pesticides	0.56
Phthalates	0.34

[a]p-value

Supplementary Table 3:

Heavy Metals: Summary Table of Kruskal – Wallis (KW) rank sum test. [b]Dunn multiple comparison post-hoc test.

		[b]Dunn multiple comparison post-hoc test					
		Between group comparisons					
Metal	[a]KW Rank Sum Test	A-B	A-C	B-C	A-D	B-D	C-D
Aluminum (Al)	0.02	-	-	-	-	0.05	0.04
Arsenic (As)	.3 x 10^{-7}	1.9 x 10^{-6}	1.3 x 10^{-6}	0.03	0.005	0.01	0.03
Cadmium (Cd)	0.0005	0.04	-	0.004	-	-	0.01
Chromium (Cr)	0.46	-	-	-	-	-	-
Iron (Fe)	1.5 x 10^{-5}	-	-	-	0.0003	-	0.0002
Mercury (Hg)	0.0008	0.006	0.001	-	0.004	-	-
Manganese (Mn)	0.88	-	-	-	-	-	-
Nickle (Ni)	0.86	-	-	-	-	-	-
Lead (Pb)	0.05	0.03	-	-	-	-	-
Selenium (Se)	0.08	-	-	-	-	-	-
Uranium (U)	0.76	-	-	-	-	-	-
Zinc (Zn)	0.0003	-	-	-	0.0003	-	0.002

[a] KW p-value

[b]Dunn multiple comparison post-hoc test

Supplementary Table 4:

Heavy Metal Summary Statistics – average and standard deviation

	Element							
Group A Fish Family	Al *n	Al Avg (SD)	As *n	As Avg (SD)	Cd *n	Cd Avg (SD)	Cr *n	Cr Avg (SD)
Acanthuridae ([a]N=7)	7	1.13 (0.56)	7	0.42 (0.13)	5	0.003 (0.002)	6	0.04 (0.04)
Scaridae (N=11)	11	2.4 (4.1)	11	0.86 (0.62)	6	0.003 (0.004)	11	0.02 (0.02)
Group B Fish Family								
Holocentridae (N=8)	8	2.4 (2.6)	8	4.1 (2)	8	0.02 (0.02)	8	0.05 (0.05)
Chanidae (N=3)	3	0.3 (0.2)	3	5.1 (1.4)	3	0.008 (0.003)	2	0.01 (0.003)
Mugilidae (N=2)	2	0.93 (0.77)	2	15.2 (4.3)	2	0.04 (0.06)	2	0.04 (0.04)
Group C Fish Family								
Serranidae (N=8)	8	2.6 (3.1)	8	2.2 (1.8)	8	0.002 (0.002)	8	0.04 (0.03)
Lethrinidae (N=5)	5	1.4 (1.7)	5	7.5 (5.3)	4	0.008 (0.01)	5	0.04 (0.03)
Mullidae (N=12)	12	4.8 (13.2)	12	8 (5.8)	12	0.4 (0.15)	12	0.04 (0.06)
Lutjanidae (N=3)	3	0.8 (0.3)	1	1.2 (1.4)	3	0.002 (0.002)	3	0.04 (0.01)
Labridae (N=2)	2	1.7 (0.6)	2	7.3 (0.4)	2	0.0015 (0.0007)	1	0.03 (0)

Haemulidae (N=1)	1	2 (0)	1	4.2 (0)	1	0.002 (0)	1	0.01 (0)
Group D								
Fish Family								
Scombridae (N=15)	15	3.3 (3.5)	15	2.9 (1.5)	15	0.007 (0.003)	10	0.02 (0.025)

Note: Results are given as the sum each of Phthalates
Compound concentrations are in mg/ kg (wet weight) = ppm
* n= the number of fish with measurable concentrations of PAHs
[a] N = the total number of fish tested

Supplementary Table 5a: Heavy Metal Summary Statistics continued

	Element					
Group A	Cu	Cu	Fe	Fe	Hg	Hg
Fish Family	*n	Avg (SD)	*n	Avg (SD)	*n	Avg (SD)
Acanthuridae ([a]N=7)	7	0.2 (0.04)	7	3.5 (1.5)	7	0.01 (0.02)
Scaridae (N=11)	11	0.2 (0.15)	11	3 (2)	11	0.026 (0.05)
Group B						
Fish Family						
Holocentridae (N=8)	8	0.2 (0.08)	8	5.6 (4.2)	8	0.11 (.012)
Chanidae (N=3)	3	0.55 (0.15)	3	6.7 (1.6)	3	0.015 (0.003)
Mugilidae (N=2)	2	0.2 (0.08)	2	3.1 (2)	2	0.02 (0.002)
Group C						
Fish Family						
Serranidae (N=8)	8	0.14 (0.03)	8	3.1 (1.8)	8	0.03 (0.01)
Lethrinidae (N=5)	5	0.19 (0.04)	5	2.4 (1.5)	5	0.07 (0.03)
Mullidae (N=12)	12	0.18 (0.04)	12	4 (1.4)	12	0.05 (0.06)
Lutjanidae (N=3)	3	0.2 (0.05)	3	3.9 (1.5)	3	0.008 (0.01)
Labridae (N=2)	2	0.2 (0.06)	2	2.4 (1.4)	2	0.05 (0.01)

Haemulidae (N=1)	1	0.2 (0)	1	2.9 (0)	1	0.02 (0)
Group D						
Fish Family						
Scombridae (N=15)	15	0.65 (0.46)	15	6.8 (2.8)	15	0.03 (0.03)

Note: Results are given as the sum each of Phthalates
Compound concentrations are in mg/ kg (wet weight) = ppm
* n= the number of fish with measurable concentrations of PAHs
[a] N = the total number of fish tested

Supplementary Table 5b: Heavy Metal Summary Statistics continued

	Element							
Group A Fish Family	Ni *n	Ni Avg (SD)	Pb *n	Pb Avg (SD)	Se *n	Se Avg (SD)	U *n	U Avg (SD)
Acanthuridae ([a]N=7).	4	0.06 (0.08)	3	0.006 (0.003)	1	0.14 (0)	6	0.0002 (0.0002)
Scaridae (N=11)	4	0.1 (0.15)	5	0.007 (0.005)	2	0.65 (0.08)	3	0.0001 (0)
Group B								
Fish Family								
Holocentridae (N=8)	2	0.02 (0.01)	3	0.009 (0.01)	7	0.37 (0.14)	6	0.0002 (0.0001)
Chanidae (N=3)	0	0	3	0.0008 (0.0003)	3	0.36 (0.06)	0	0
Mugilidae (N=2)	0	0	2	0.0007 (0.0003)	1	0.17 (0)	1	0.0002
Group C								
Fish Family								
Serranidae (N=8)	4	0.02 (0.02)	5	0.003 (0.003)	8	0.36 (0.08)	6	0.0003 (0.0002)
Lethrinidae (N=5)	3	0.025 (0.02)	2	0.01 (0.1)	5	0.33 (0.04)	1	0.0002
Mullidae (N=12)	5	0.06 (0.04)	8	0.006 (0.01)	5	0.3 (16)	3	0.0001

Lutjanidae (N=3)		1	0.008	2	0.003 (0.0006)	1	0.4 (0)	1	0.0003
Labridae (N=2)		0	0	2	0.004 (0.002)	2	0.3 (0.04)	1	0.0001
Haemulidae (N=1)		0	0	1	0.005 (0)	0	0	0	0
Group D									
Fish Family									
Scombridae (N=15)	15	5	0.04 (0.06)	14	0.009 (0.02)	15	0.35 (0.18)	7.	0.0001 (0.0001)

Note: Results are given as the sum each of Phthalates

Compound concentrations are in mg/ kg (wet weight) = ppm

* n= the number of fish with measurable concentrations of PAHs

[a] N = the total number of fish tested

Supplementary Table 5c: Heavy Metal Summary Statistics continued

	Element	
Group A	Zn	Zn
Fish Family	*n	Avg (SD)
Acanthuridae ([a]N=7)	7	3.1 (0.8)
Scaridae (N=11)	11	2.9 (1.4)
Group B		
Fish Family		
Holocentridae (N=8)	8	3.3 (1.5)
Chanidae (N=3)	3	6.1 (1.4)
Mugilidae (N=2)	2	3.1 (0.9)
Group C		
Fish Family		
Serranidae (N=8)	8	3.4 (1.
Lethrinidae (N=5)	5	3.1 (2)
Mullidae (N=12)	12	3.4 (0.99)
Lutjanidae (N=3)	1	3.2 (0.2)
Labridae (N=2)	2	2.7 (0.4)
Haemulidae (N=1)	1	3.6 (0)

Group D		
Fish Family		
Scombridae (N=15)	15	5 (1.1)

Note: Results are given as the sum each of Phthalates

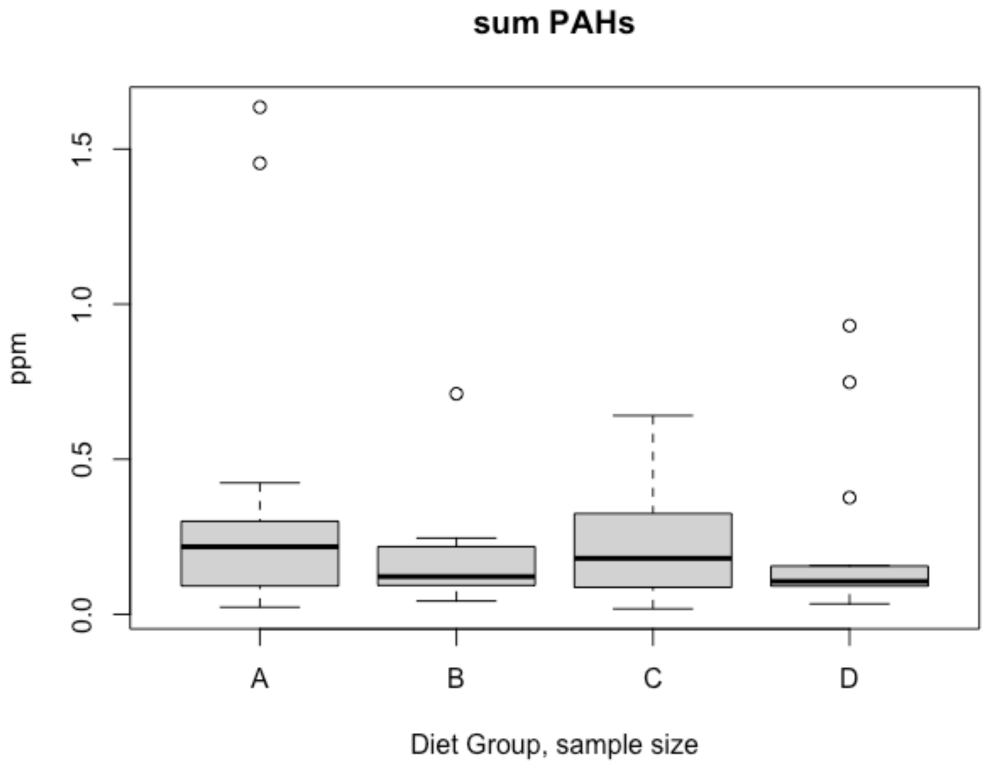
Compound concentrations are in mg/ kg (wet weight) = ppm

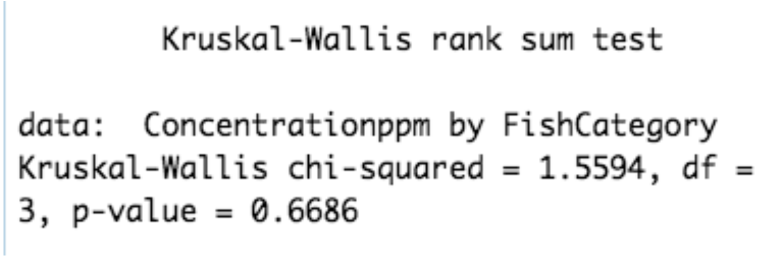
* n= the number of fish with measurable concentrations of PAHs

[a] N = the total number of fish teste

Chapter 2
Non-significant graphical analyses
Organic Contaminants

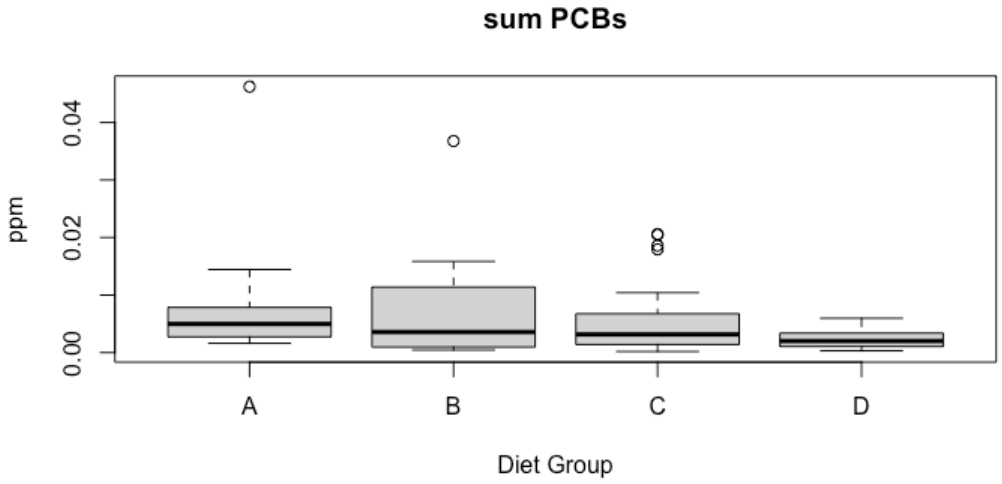
PAHs

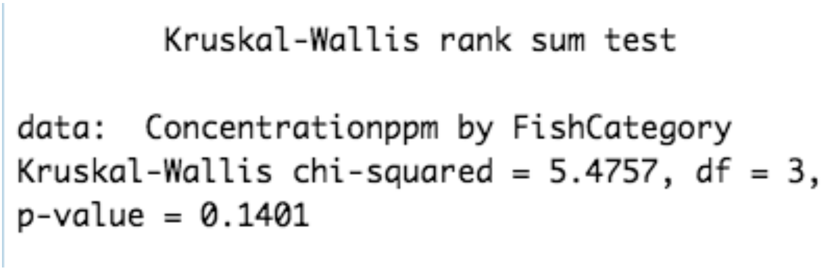




```
        Kruskal-Wallis rank sum test

data:  Concentrationppm by FishCategory
Kruskal-Wallis chi-squared = 1.5594, df =
3, p-value = 0.6686
```

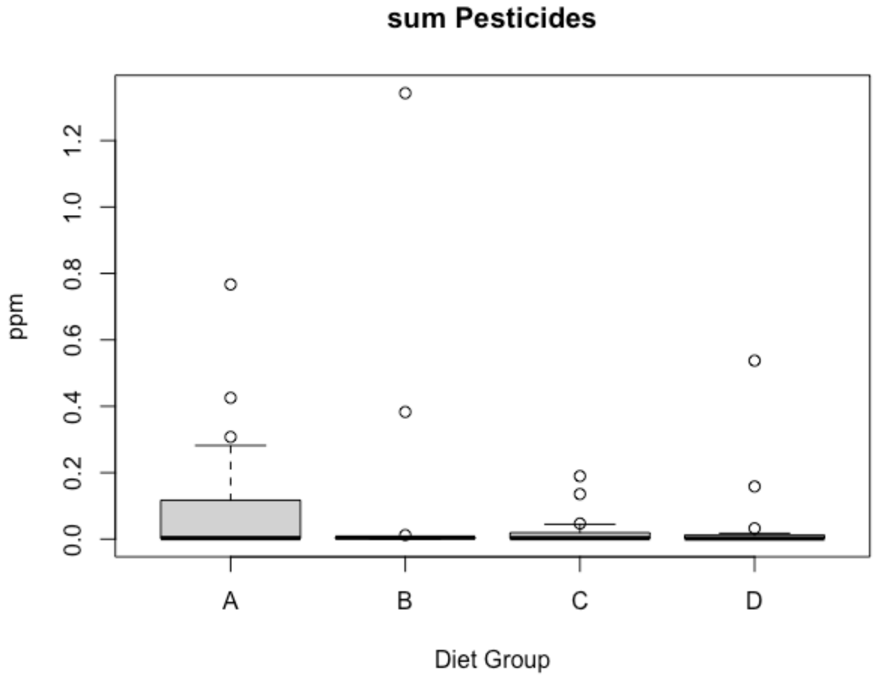
PCBs

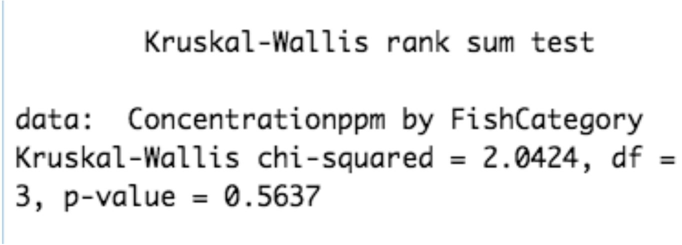




```
        Kruskal-Wallis rank sum test

data:  Concentrationppm by FishCategory
Kruskal-Wallis chi-squared = 5.4757, df = 3,
p-value = 0.1401
```

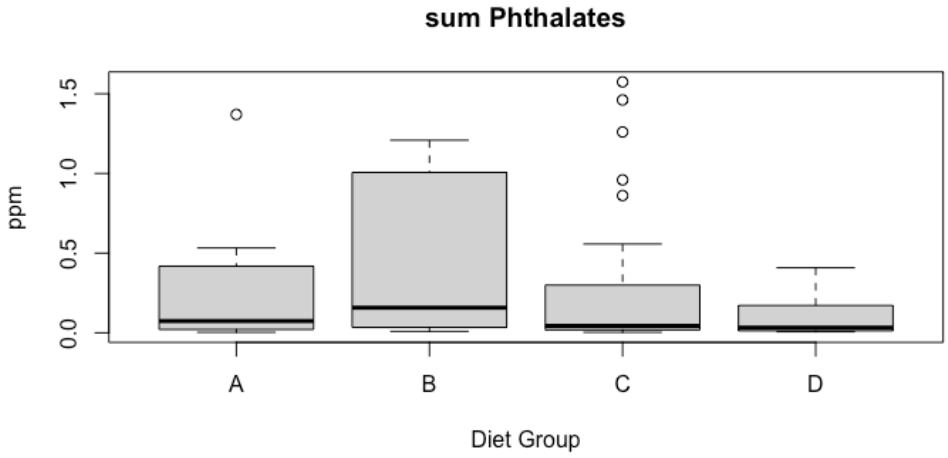
Pesticides

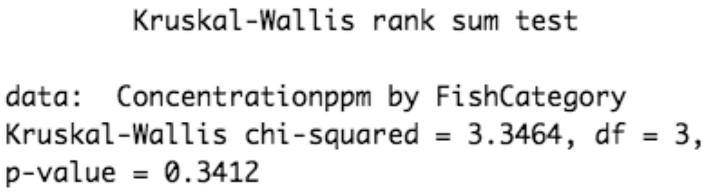




```
        Kruskal-Wallis rank sum test

data:  Concentrationppm by FishCategory
Kruskal-Wallis chi-squared = 2.0424, df =
3, p-value = 0.5637
```

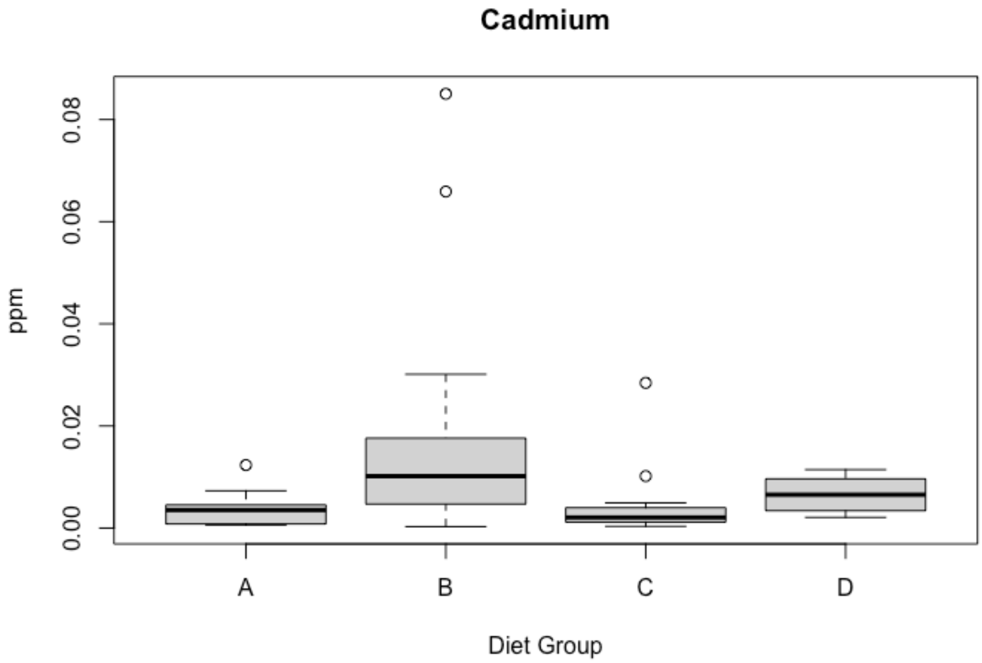
Phthalates

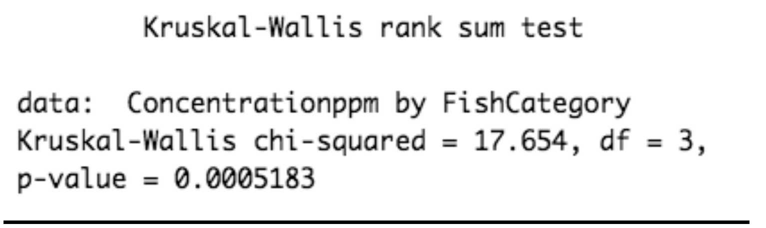




```
        Kruskal-Wallis rank sum test

data:  Concentrationppm by FishCategory
Kruskal-Wallis chi-squared = 3.3464, df = 3,
p-value = 0.3412
```

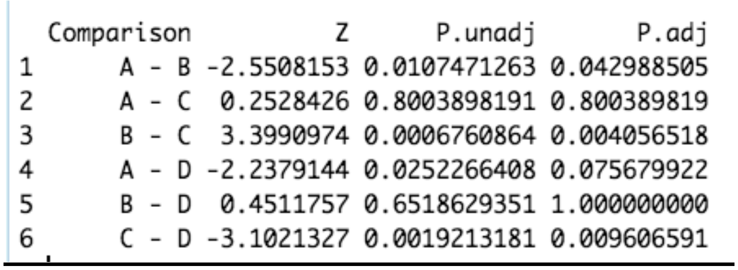
Heavy Metals
Cadmium





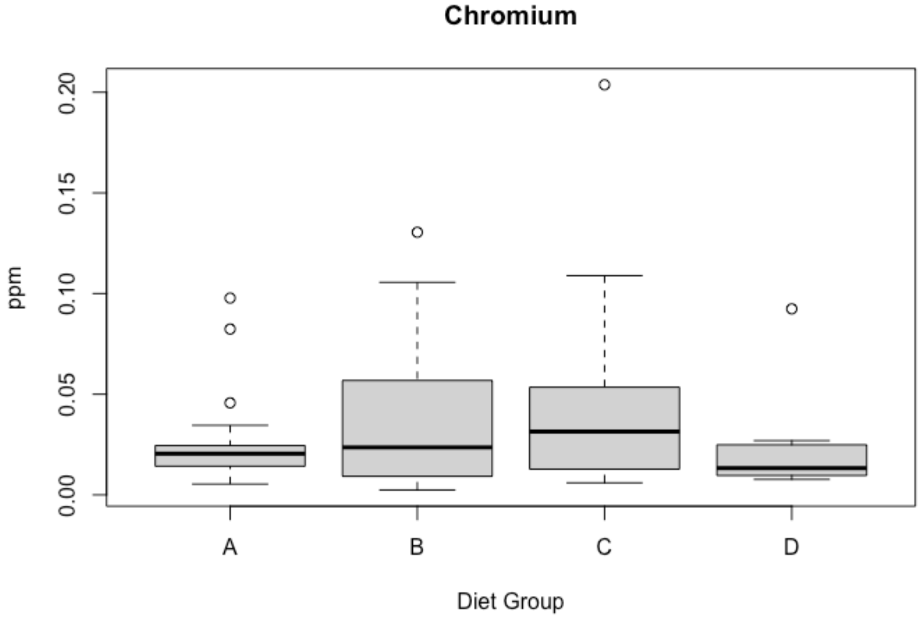
```
	Kruskal-Wallis rank sum test

data:  Concentrationppm by FishCategory
Kruskal-Wallis chi-squared = 17.654, df = 3,
p-value = 0.0005183
```

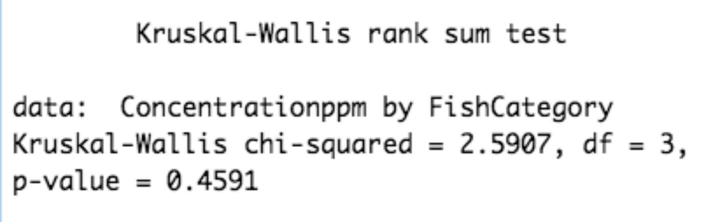
Dunn's post-hoc test



```
  Comparison          Z      P.unadj       P.adj
1      A - B -2.5508153 0.0107471263 0.042988505
2      A - C  0.2528426 0.8003898191 0.800389819
3      B - C  3.3990974 0.0006760864 0.004056518
4      A - D -2.2379144 0.0252266408 0.075679922
5      B - D  0.4511757 0.6518629351 1.000000000
6      C - D -3.1021327 0.0019213181 0.009606591
```

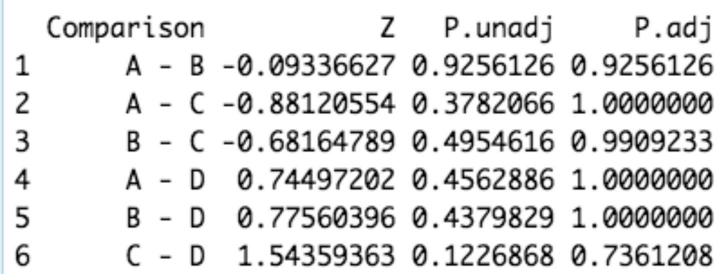
Chromium





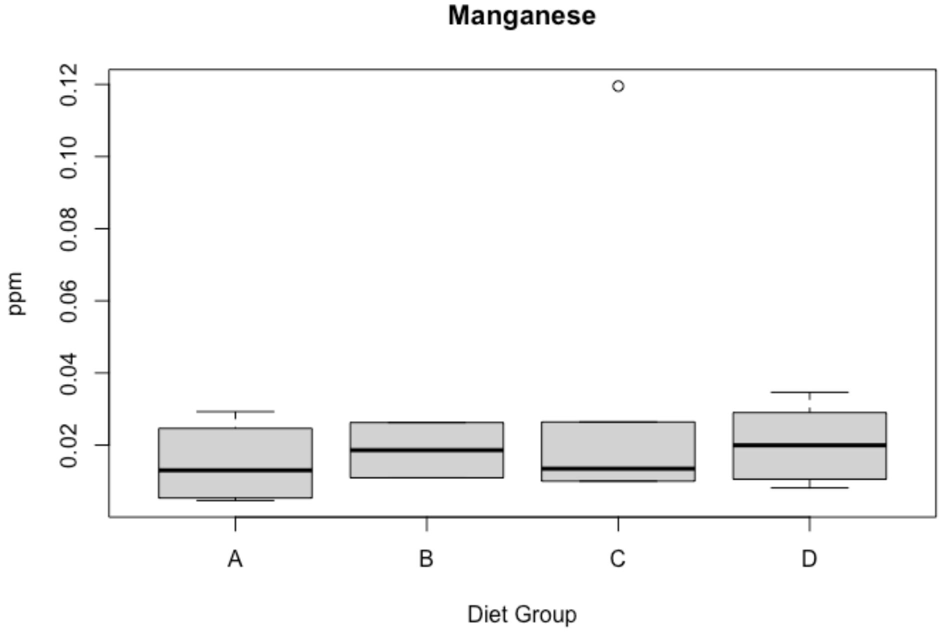
```
        Kruskal-Wallis rank sum test

data:  Concentrationppm by FishCategory
Kruskal-Wallis chi-squared = 2.5907, df = 3,
p-value = 0.4591
```

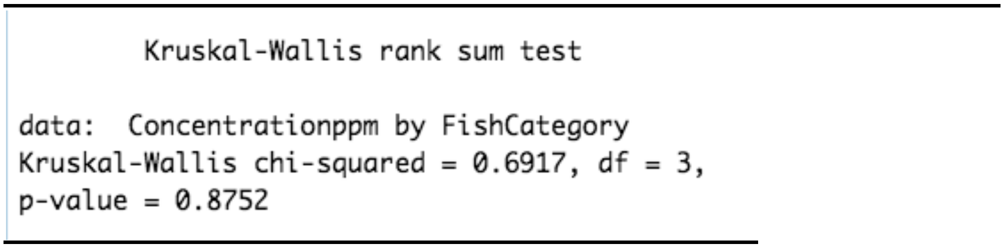
Dunn’s post-hoc test



```
  Comparison           Z   P.unadj     P.adj
1      A - B -0.09336627 0.9256126 0.9256126
2      A - C -0.88120554 0.3782066 1.0000000
3      B - C -0.68164789 0.4954616 0.9909233
4      A - D  0.74497202 0.4562886 1.0000000
5      B - D  0.77560396 0.4379829 1.0000000
6      C - D  1.54359363 0.1226868 0.7361208
```

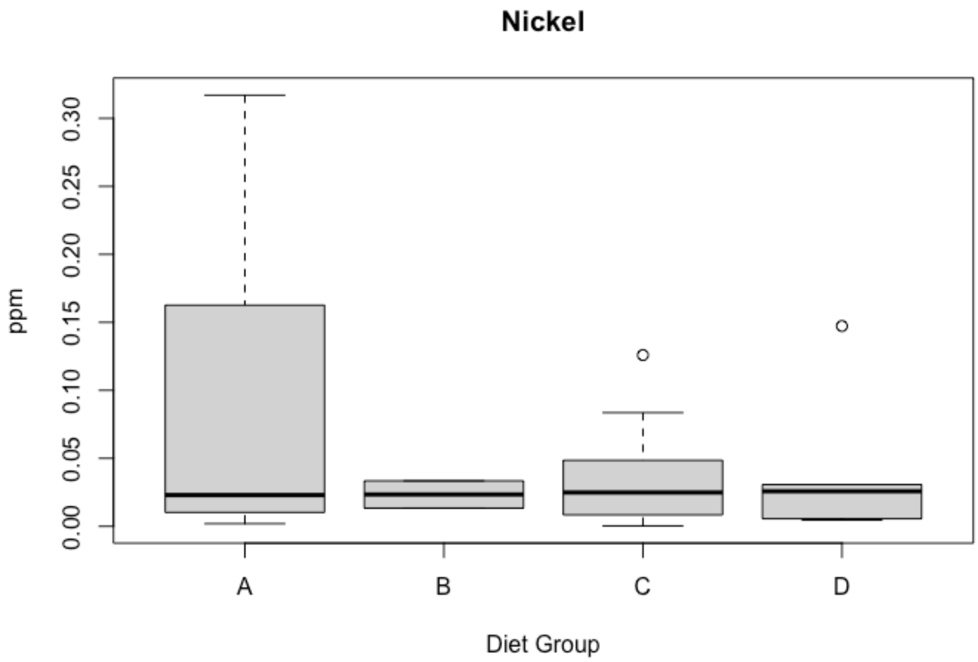
Manganese

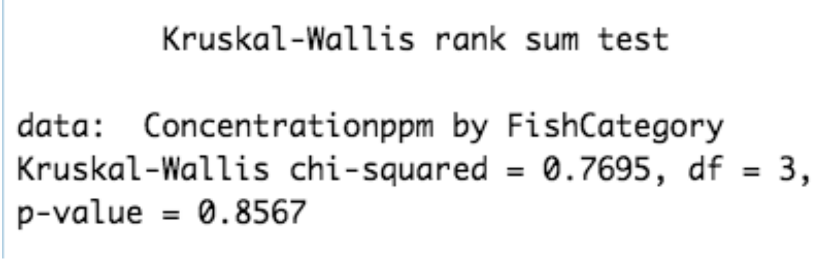




```
        Kruskal-Wallis rank sum test

data:  Concentrationppm by FishCategory
Kruskal-Wallis chi-squared = 0.6917, df = 3,
p-value = 0.8752
```

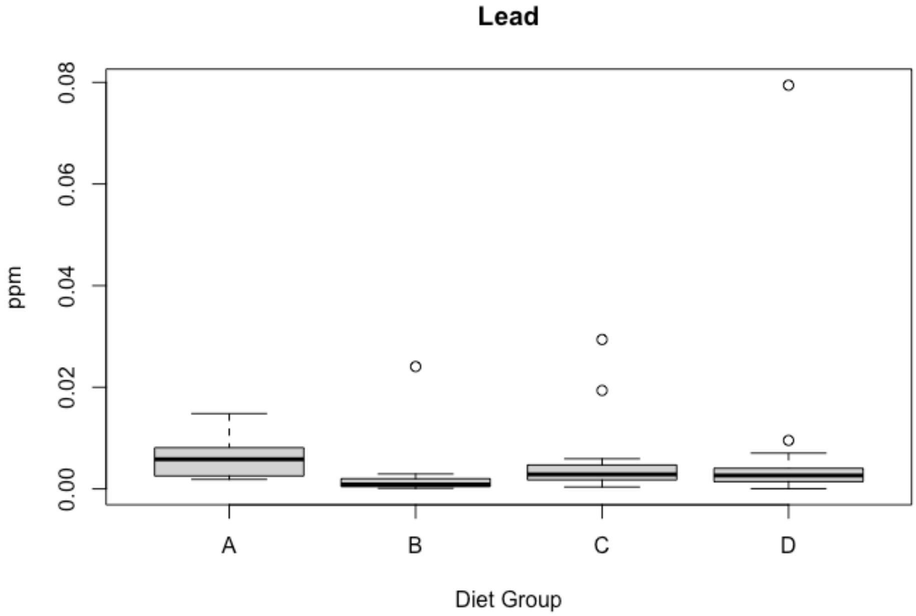
Nickel

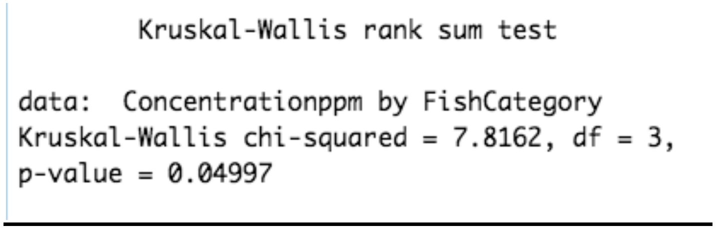




```
        Kruskal-Wallis rank sum test

data:  Concentrationppm by FishCategory
Kruskal-Wallis chi-squared = 0.7695, df = 3,
p-value = 0.8567
```

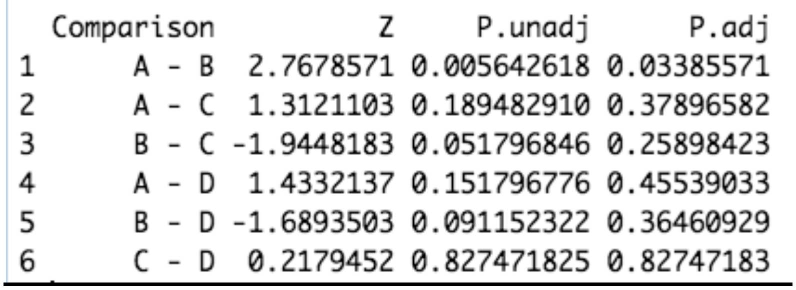
Lead





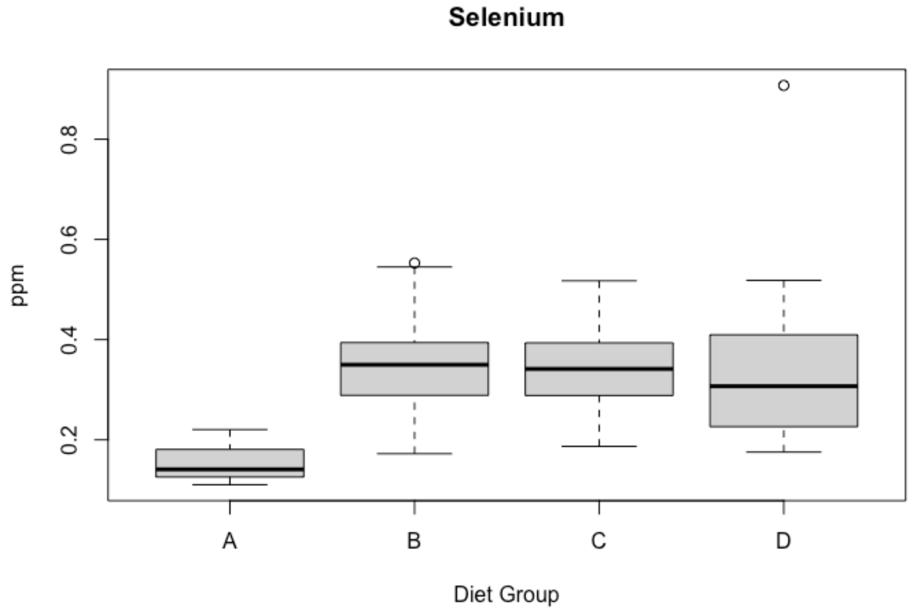
```
        Kruskal-Wallis rank sum test

data:  Concentrationppm by FishCategory
Kruskal-Wallis chi-squared = 7.8162, df = 3,
p-value = 0.04997
```

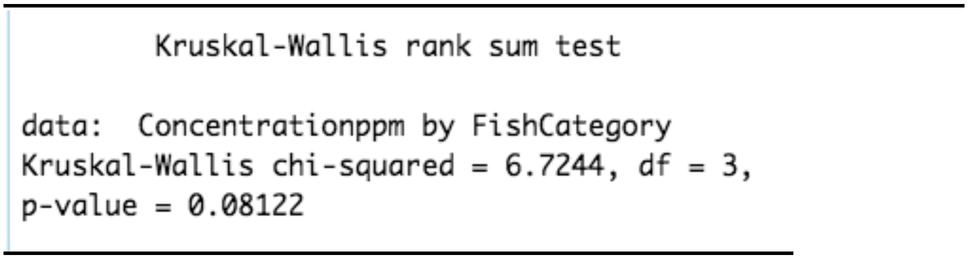
Dunn's post-hoc test



```
  Comparison          Z     P.unadj      P.adj
1      A - B  2.7678571 0.005642618 0.03385571
2      A - C  1.3121103 0.189482910 0.37896582
3      B - C -1.9448183 0.051796846 0.25898423
4      A - D  1.4332137 0.151796776 0.45539033
5      B - D -1.6893503 0.091152322 0.36460929
6      C - D  0.2179452 0.827471825 0.82747183
```

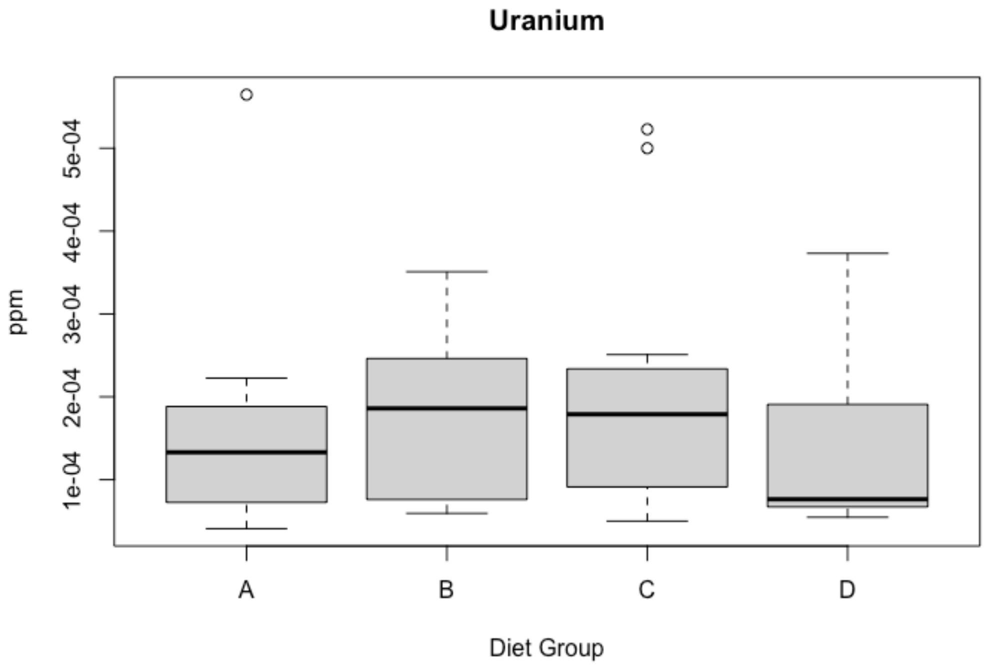
Selenium

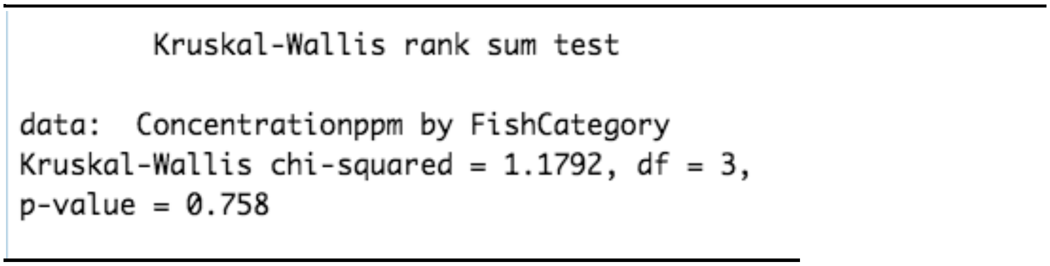




```
	Kruskal-Wallis rank sum test

data:  Concentrationppm by FishCategory
Kruskal-Wallis chi-squared = 6.7244, df = 3,
p-value = 0.08122
```

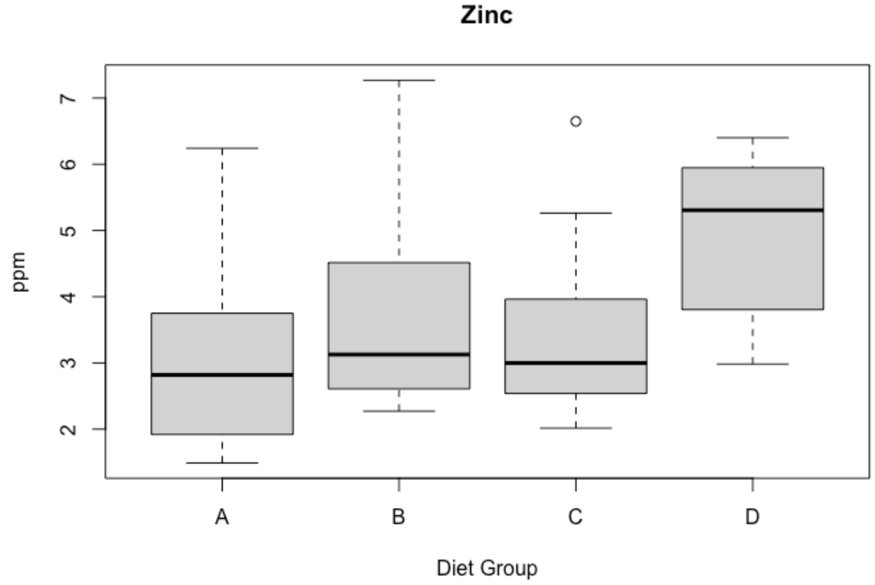
Uranium

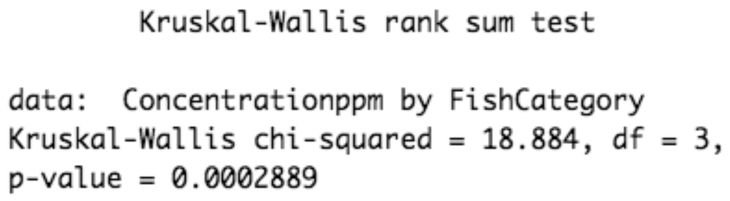




```
        Kruskal-Wallis rank sum test

data:  Concentrationppm by FishCategory
Kruskal-Wallis chi-squared = 1.1792, df = 3,
p-value = 0.758
```

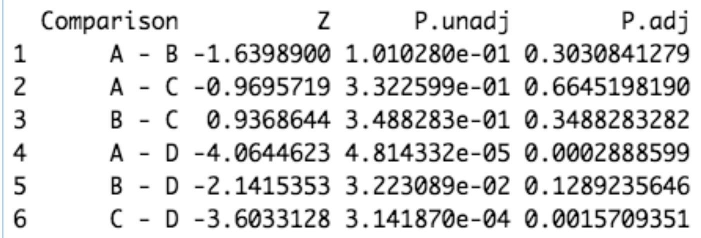
Zinc





```
        Kruskal-Wallis rank sum test

data:  Concentrationppm by FishCategory
Kruskal-Wallis chi-squared = 18.884, df = 3,
p-value = 0.0002889
```

Dunn’s post-hoc test



```
  Comparison          Z      P.unadj        P.adj
1      A - B -1.6398900 1.010280e-01 0.3030841279
2      A - C -0.9695719 3.322599e-01 0.6645198190
3      B - C  0.9368644 3.488283e-01 0.3488283282
4      A - D -4.0644623 4.814332e-05 0.0002888599
5      B - D -2.1415353 3.223089e-02 0.1289235646
6      C - D -3.6033128 3.141870e-04 0.0015709351
```

APPENDIX B

SUPPORTING INFORMATION FOR CHAPTER 3

Chapter 3: Canned Meats

American Samoa Personalized Recruitment Script
I am conducting a survey to better understand how much seafood and pisupo American Samoans eat. I will use this information, in conjunction with the contaminant analyses (ie pesticides, etc.) of the food, to evaluate if there is any risk to your health associated with eating these items. May I please ask you a few questions related your household seafood and pisupo intake? Your personal identify information will remain confidential. I am interested in creating accurate generalizations about American Samoans as a whole, not about your family in particular. The survey (oral or written) is anticipated to take approximately 30 minutes. Please complete as much of it as you can and/ or are willing to. I appreciate your time.
Your participation in this survey is voluntary. If you have any questions, please feel free to email me at ecotlewis@asu.edu.

Survey response is limited to resident adult humans of American Samoan descent

2. Date:

<u>Personal Information</u>

3. Age:
4. Gender:
5. Village:
6. How many people are in your home? What are their ages?

<u>Fish consumption data:</u>

   1) Do you consume locally caught fish?
   2) Which type of fish do you consume?
   3) raw? Cooked?
   4) Have you eaten any fish or shellfish in the last 24 hours? How much?
   5) How many palm sized pieces per week? (I chose per week because that is how mercury accumulation is assessed).
   6) Where do you get it from? (Catch or purchase? (Source info.))
   7) Do YOU have a fishing license?
   8) Do you consume the same amount of fish all throughout the year? If not, how does it change?
   9) Why do you or don't you consume fish?

<u>Palolo consumption data:</u>

   1) Do you consume palolo?
   2) How do you eat it?
   3) How much?
   4) Do you consume only at the event? Year round?
   5) Why do you or don't you consume palolo?

<u>Shellfish consumption data:</u>

   1) Do you consume shellfish? Which type?
   2) Where do you get your shellfish from?
   3) How many individuals do you eat per meal?
   4) How many meals per week?

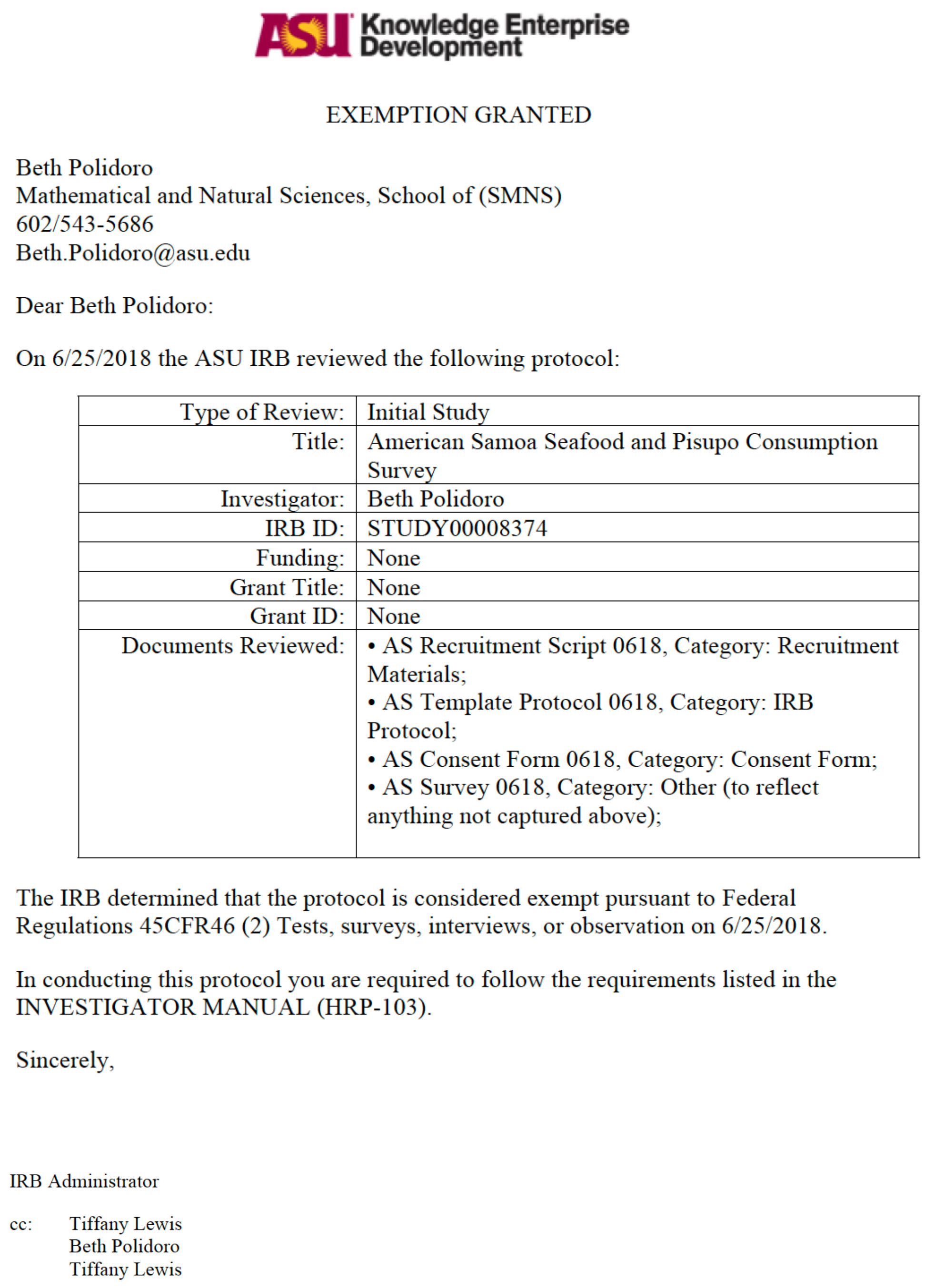
5) Why do you or don’t you consume shellfish?
6) How does this compare to your families’ historic consumption of local shellfish? Please tell me any memories you have of local shellfish, either firsthand or legend.

Pisupo consumption data:

1) Do you consume pisupo? Which brand?
2) How much of the can do you eat in one sitting?
3) How many meals per week?
4) Does your family consume more or less pisupo than you do? How about the kids? How much?
5) Why do you or don’t you consume pisupo?

Do you have any favorite or fish, seafood, shellfish, or pisupo recipes that you’d like to share?

Survey IRB Approval



ASU Knowledge Enterprise Development

EXEMPTION GRANTED

Beth Polidoro
Mathematical and Natural Sciences, School of (SMNS)
602/543-5686
Beth.Polidoro@asu.edu

Dear Beth Polidoro:

On 6/25/2018 the ASU IRB reviewed the following protocol:

Type of Review:	Initial Study
Title:	American Samoa Seafood and Pisupo Consumption Survey
Investigator:	Beth Polidoro
IRB ID:	STUDY00008374
Funding:	None
Grant Title:	None
Grant ID:	None
Documents Reviewed:	• AS Recruitment Script 0618, Category: Recruitment Materials; • AS Template Protocol 0618, Category: IRB Protocol; • AS Consent Form 0618, Category: Consent Form; • AS Survey 0618, Category: Other (to reflect anything not captured above);

The IRB determined that the protocol is considered exempt pursuant to Federal Regulations 45CFR46 (2) Tests, surveys, interviews, or observation on 6/25/2018.

In conducting this protocol you are required to follow the requirements listed in the INVESTIGATOR MANUAL (HRP-103).

Sincerely,

IRB Administrator

cc: Tiffany Lewis
Beth Polidoro
Tiffany Lewis