

**SEDIMENT QUALITY IN FISHERMAN BAY
AND FRIDAY HARBOR, WA**

**PETROLEUM RESIDUES, POLYCYCLIC AROMATIC
HYDROCARBONS, PYRETHROID PESTICIDES, AND
TOXIC METALS**



**Russel Barsh
Courtney Clark
Tiffany Stephens**

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KWIÀHT

Center for the Historical Ecology of the Salish Sea
PO Box 415, Lopez, WA 98261

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SUMMARY

Marine sediment grabs and cores were collected along transects across Fisherman Bay and Friday Harbor, two San Juan County working waterfronts associated with Urban Growth Areas. Sediments were sieved to determine the proportions of sand and silts, and extracted in organic solvents for measurement of Total Petroleum Hydrocarbons (TPH), low-weight Polycyclic Aromatic Hydrocarbons (PAHs), copper and zinc (attributable to anti-fouling paints), and over-the-counter pyrethroid pesticides, all predicted to be part of the chemical footprint of island roads, boats, and homes.

Sediment TPH and PAH concentrations were three times greater in Friday Harbor than Fisherman Bay, reflecting the relative size of the two towns and their marinas; while copper and zinc were somewhat more concentrated in Fisherman Bay. Concentrations of PAHs, pyrethroids, and metals were all at threshold levels of probable biological effects, indicating a need to manage runoff in these two Urban Growth Areas more effectively.

As San Juan County's Urban Growth Areas continue to build and grow, managing and monitoring contaminants in bay sediments will grow in importance, lest development undermine the integrity of the marine ecosystems on which much of the San Juan Islands' popularity, prosperity, and local seafood resources depend.

We recommend a more intensive baseline study of sediment contamination in all San Juan County working waterfront, and monitoring including model organisms such as mussels or clams as well as sediments.

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The San Juan Islands have experienced very rapid population growth and build-out since the 1970s, including roads perched on shoreline bluffs, shoreline armoring, docks, and boat traffic. Much of this growth has been concentrated in four working bays: Friday Harbor (San Juan Island), Eastsound (Orcas), Fisherman Bay (Lopez), and Roche Harbor (San Juan Island). Friday Harbor, Eastsound, and the “village” on Fisherman Bay are designated Urban Growth Areas (UGAs) subject to State growth-planning standards. All four working bays are also subject to the State’s Shoreline Management Act and more recent storm water (drainage and surface runoff) standards.

General principles of hydrogeology, geomorphology, and toxicology predict that the hardening of surfaces and shorelines, proliferation of docks, and increase in road use and boat traffic, has made the islands’ four working bays siltier and more contaminated—in particular, contaminated with toxics associated with paving materials such as asphalts, creosote and metals used to preserve pilings, metals used in anti-fouling boat paints, and internal-combustion engine fuels, lubricants, and exhaust. Chief amongst these concerns would ordinarily be polycyclic aromatic hydrocarbons (PAHs), carcinogenic constituents of petroleum, incompletely burnt fossil fuels and oils, creosote, asphalt and bitumen; and toxic metals used in marine paints and preservatives such as copper and zinc, which have proven to be exceptionally toxic to fish.

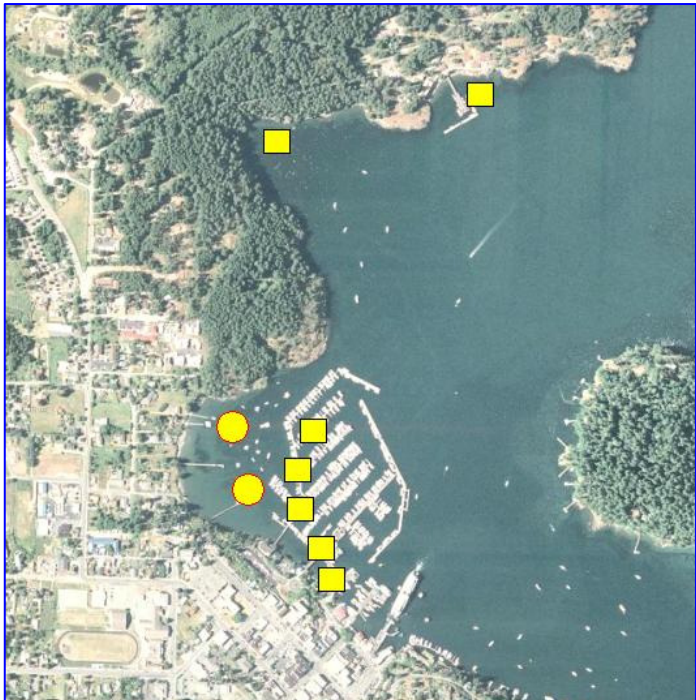
Reliable data on actual current levels of silt and contaminants in the islands’ bays would be useful for environmental monitoring and the management of the “footprint” of future development, such as shoreline infrastructure, erosion controls in the face of rising sea levels, and urban storm sewer discharges. Baseline data is also useful in the event of a future spill or other accident. Absent a reliable baseline, the marginal contribution of a spill to sediment contaminant loads cannot be determined

Relatively little is known about historical changes in sediments in working bays of San Juan County, however. To address this gap, we designed the present pilot study in cooperation with the San Juan Nature Institute and local schools, and with the support of The Russell Family Foundation. Elements of the study were assigned to middle and high school students as field and classroom teaching exercises.

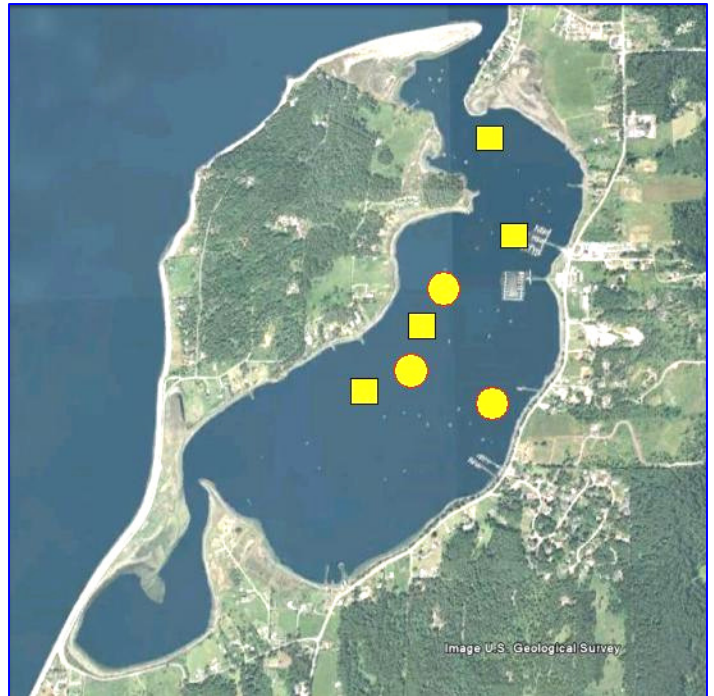
Methods

Previous research had found significant differences in the bulk toxicity of shallow sediments from Fisherman Bay and Friday Harbor, which were hypothesized to represent different levels of tidal flushing—comparatively low in Fisherman Bay—and dispersal or dilution of toxicants (Barsh 2009a). Accordingly, we re-visited these two bays to collect a larger number of surface grabs as well as some shallow cores, and investigate sediment structure and several specific adsorbed contaminants.

Maps 1 and 2 identify the locations of the grabs and cores collected in April 2010. Cores (circles) were taken where we expected the lowest circulation and highest rates of sediment accumulation; grabs (squares) were used to obtain a more representative cross-section of conditions throughout the bay. A hand-deployed AMS bottom dredge was used to collect grabs working from docks and a small boat. A 20-inch stainless steel Wildco™ torpedo-type coring tool, mounted on 15 feet of steel drive rods, was deployed by hand over the side of a small boat.



Map 1: Friday Harbor sampling sites



Map 2: Fisherman Bay sampling sites

All coring encountered firm compact sands at relatively shallow depths, averaging 15 cm in Friday Harbor and 22 cm in Fisherman Bay. We hypothesized that these sands represented pre-development conditions 40-50 years ago. Accretion rates of 0.5 cm/year would not have been unreasonable during this period. Small but measurable quantities of lead in the deepest and oldest slices of our cores are consistent with this hypothesis, since leaded gasoline was being phased out of use by the 1980s.

After collection, cores were sliced and the slices sub-sampled. After scraping any detritus (mainly decaying diatoms) from the top of each core, sediment was scooped from the top 2 cm, the bottom 2 cm, and for the longer Fisherman Bay cores, the middle 2 cm. Sterile conical tubes were used to remove 1-cc sub-samples of each slice for bacteriology. An aluminum spoon was then used to take two approximately 20-gram sub-samples from each slice for other purposes. One 20-gram sub-sample was immediately dispersed in 50 mL of HPLC-grade methanol and left overnight before filtering 5 mL of liquid through a 0.22-micron syringe filter for PAH testing. The solids and remaining liquid were then acidified with 5 mL of 15% hydrochloric acid, shaken, allowed to settle for one hour, and

filtered through a 0.22-micron syringe filter to extract metal ions fully into the methanol. Solids were then dried and weighed.

The other 20-gram sub-sample was dried at ambient temperature, then suspended in 15 mL of hexane and sonicated for 10 minutes. After the solids settled, all supernatant liquid was poured into pre-weighed Nalgene tubes and allowed to evaporate at ambient temperature until less than 1 mL liquid remained. To complete the evaporation of hexane and any moisture, tubes were placed in a water bath at 60°C for approximately one hour. Tubes were then cooled and weighed. The difference in tube weight, adjusted for the dry weight of the extracted solids, was recorded as TPH.

Dried hexane-extracted solids were ground in a ceramic mortar and separated into size classes using a Keck sand shaker. Each fraction was weighed, and examined under a microscope for grain shape and mineralogy. Sediments smaller than 0.25 mm are treated as “silts” and tend to be composed largely of clay minerals.

Acidified methanol extracts were tested colorimetrically for copper and zinc using the Diethyldithiocarbamate (DDC) and Zincon standard methods, respectively. Reagent blanks and controls indicate mean measurement errors of 0.5 ppm for copper and 0.75 for zinc. We used ELISA immunoassays to screen methanolic extracts for PAHs (Strategic Diagnostics, Newark, DE) and pyrethroid pesticides (Abraxis LLC, Warminster, PA). In both immunoassays, results for positive controls were accurate within 20 percent. ELISA data should nevertheless be treated as provisional until confirmed by mass spectrometry.

Results

All of the Fisherman Bay sediments were composed of sharp, prismatic, mainly clear quartz grains with a small number of prismatic black hornblende grains and mica flakes; and finely divided gray clay minerals; matching the Whiskey Hill moraine on the east side of the bay. Rounded grains, indicative of long-distance water transport or beach residence, were absent. This suggests that the Fisherman Bay sediments were transported to the bay by short-distance surface runoff.

Friday Harbor sediments were more rounded and included a larger proportion of opaque “milky” quartz, matching the few remaining low bluffs around the bay, composed of glacial outwash. No evidence of historical changes in sediment sources were observed in either bay.

Table 1 summarizes the grain size analysis of Fisherman Bay sediments including four surface grabs as well as three 20-25 cm deep cores. Grabs varied greatly in siltiness, depending on their location within the bay. The coarsest grab was collected closest to the ship channel leading north out of the bay, where tidal exchanges are strongest.

As predicted, there has been an increase in siltiness of sediments. The bottoms of all three cores contained coarser material than the more recent middle and top slices. But the trend was not strong: a 12 percent increase in siltiness in core I over an estimated time period of 30-50 years, 9 percent in core II, and 5 percent in core III. A longer historical record from deeper cores might identify an older, stronger shift in sediment size.

Table 2 summarizes the grain size analysis of Friday Harbor sediments, including two surface grabs and two cores. Recent sediments were predictably a little coarser than Fisherman Bay, and there was some evidence of increasing siltiness over time, especially in core B collected closer to the armored shore. Major changes in the two bays may have occurred earlier (deeper) than we were able to reach with our coring tools.

Sediment contaminant results are summarized in Tables 3 and 4.

Total Petroleum Hydrocarbons (TPH) was consistently higher in Friday Harbor at an average of 3.6 percent of sediments by weight, than in Fisherman Bay at 1.2 percent of sediments by weight—suggesting that the larger scale of petroleum inputs from roads and boating to Friday Harbor, offsets Friday Harbor’s better tidal circulation. The data imply that petroleum inputs to Friday Harbor are more than three times the inputs to Fisherman Bay, which is not implausible. One sample from each bay appears anomalously low, but in actuality strengthens our analysis. Grab 2 in Fisherman Bay was taken from just inside the channel and was mainly coarse sand. Grab D was taken from the docks at the Friday Harbor Marine Laboratories, nearly in San Juan Channel. These two sites enjoy stronger tidal circulation than any of our other sampling sites.

PAHs, as some toxic components of petroleum, were also about three times more concentrated in sediments from Friday Harbor than sediments from Fisherman Bay: 0.79 parts per million on average, compared with 0.25 parts per million. Our measurements of TPH and PAHs are consistent, indicating that they both arise from the same sources, and represent considerably greater inputs to Friday Harbor from road runoff, docks and boats.

Copper and zinc, highly toxic to fish and other aquatic animals, are very scarce in San Juan County surface waters, in our experience rarely exceeding 0.5 parts per million. Both metals are widely used in marine anti-fouling treatments, and we expected that they would be enriched markedly in busy harbors. We found significant levels of both copper and zinc in both bays studied as predicted. Concentrations of these metals were higher in Fisherman Bay, however. It is not possible to determine whether this is simply a result of poor circulation, or also reflects disproportionately large inputs.

Pyrethroid pesticides are markers for runoff from homes and gardens. We were only able to assay pyrethroids in sediments from Fisherman Bay, where concentrations averaged 0.3 parts per million (300 parts per billion), comparable to levels we previously recorded in Eastsound bivalves (Barsh et al. 2009b). Higher levels are likely to be found in Fisherman Bay bivalves and other marine organisms due to bioaccumulation.

No clear trends in levels of organic pollution emerge from our data; the number of cores was relatively small, and variance in ELISA results relatively great. A large sample of deeper cores may produce more cogent results. Nevertheless, TPH and copper clearly increased over time in all but one of the cores (Table 5), and zinc increased in three out of five cores. All cores showed an increase the proportion of silt.

The coarsest sediments (for example Fisherman Bay grab 2) were also “cleanest,” as predicted from the relationship between adsorption and grain size. Subdividing solids into smaller particles increases total surface area available for adsorption. Increased silts therefore should in principle result in higher concentrations of contaminants—unless the inputs have meanwhile decreased. Decreased inputs due to re-engineering of the Port of

Friday Harbor may help explain why some Friday Harbor sediment cores were “cleaner” at the top than the older middle or bottom.

Discussion

Increasing silt ratios reflect an increasing capacity of bay sediments to accumulate toxic contaminants. They presumably result from an increase in the paved, impermeable surface area and shoreline armoring of the Fisherman Bay and Friday Harbor watersheds; combined, in the case of Fisherman Bay, with decreased tidal circulation caused by the construction of Bayshore Road on top of the tombolo. Build-out, population growth, and increased consumption of oils, fuels, and outdoor products are synergistic, increasing the adsorptive capacity of sediments as well as inputs of toxic compounds. Despite efforts to manage growth in San Juan County the marine chemical footprints of Fisherman Bay and Friday Harbor have been growing.

Increasing sediment TPH levels are one indicator of the growing chemical loading of San Juan County bays. Sediment TPH is easy to measure, and variance was relatively low among samples we collected from the same bay, making it promising for monitoring future impacts of build-out, growth and runoff management in the islands. TPH data will also be useful as a baseline in the unfortunate event of a future marine fuel spill. TPH is not a measure of toxicity, however. But some of the hundreds of chemical constituents of petroleum can be highly toxic, and can be measured directly, such as PAHs.

Low-weight PAHs such as naphthalene, phenanthrene, and benzo- α -pyrene (BaP) have long been identified as carcinogens at part-per-billion levels. Asphalt plants, asphalt road paving operations, and vehicle emissions are the leading sources of human exposure to PAHs (Centers for Disease Control 2005). Road paving sources include the seal-coats commonly applied to San Juan County roads (Mahler et al. 2005). Road runoff transports PAHs from paving materials and vehicles to aquatic and marine environments. Creosote from treated pilings is also a significant source of PAHs in marine habitats (Smith 2008). PAH concentrations vary considerably in these different source materials, thus should not simply be assumed to be a predictable fraction of TPH levels.

Our immunoassays of sediments nevertheless found that PAH levels, like TPH, were three times greater in Friday Harbor than in Fisherman Bay. Concentrations in both bays exceeded the EPA primary drinking water standard of 0.2 parts-per-billion: they are high enough to pose a risk to human health, if directly ingested, but more likely of course to affect humans and other mammals, such as orcas, otters and seals, that ingest seafoods. PAHs are not very soluble in water, but sediment scavengers and suspension feeders such as infanal worms and clams move sediment PAHs into the marine food web, where they bioaccumulate (McFarland 1998). Without additional measurements, we cannot compute the rate of uptake of PAHs by the marine organisms in Friday Harbor or Fisherman Bay; but their tissue concentrations are almost certainly at least equal to the concentrations we observed in bay sediments.

Copper and zinc, by comparison, are more concentrated in Fisherman Bay, which may reflect a combination of greater use in boat maintenance and repair activity, and very poor tidal circulation. Metals in sediments are not completely bioavailable and as little as

one percent may actually be accumulate in fish. Current sediment levels in the range of 30-50 part-per-million represent uptake levels as little as 0.5 parts-per-million. One part-per-million of copper can be toxic to fish, however.

Most household and garden pesticides sold in San Juan County contain pyrethroid pesticides, as do the products preferred by the county's pest-control operators. Waxy and poorly soluble in water, pyrethroids are rendered mobile and sprayable by the addition of surfactants. Applied outdoors, pyrethroids can easily be transported to marine habitats by rain runoff, either suspended in water by the surfactants with which they are packaged, or adsorbed to particles of silt.

Pyrethroids affect respiration, olfaction, and disrupt endocrine signaling in Pacific salmon (Tollefson et al. 2002; Wheelock et al. 2005; Tierney et al. 2006). Concentrations as low as 0.1 parts per billion can be toxic to rainbow trout (*Oncorhynchus mykiss*) (Todd et al. 2003). Pyrethroids also pose a threat to marine invertebrates on which fish feed.

Pyrethroid pesticide concentrations in Fisherman Bay sediments are comparable to concentrations previously reported in Eastsound bivalves (Barsh et al. 2009b). Federal pesticide-residue standards prohibit the sale of meat or eggs with over 50 parts per billion of the pyrethroid pesticide deltamethrin (Environmental Protection Agency 2004), which is one-sixth of the pyrethroid concentration we measured in Fisherman Bay sediments. It should be borne in mind that pyrethroid pesticides adsorbed to fine sediments may not all be bioavailable; concentrations in shellfish and fish may be lower.

Recommendations

Our study found toxic chemicals associated with roads, cars and boats, homes and gardens, at levels of probable biological effects in two of San Juan County working bays, Friday Harbor and Fisherman Bay. As San Juan County's Urban Growth Areas continue to build and grow, managing and monitoring contaminants in bay sediments will grow in importance, lest development undermine the integrity of the marine ecosystems on which much of the islands' popularity, prosperity, and local seafood resources depend.

A first step is fully documented baseline evaluation of local source contaminants in sediments from all of the county's working harbors including Eastsound, Deer Harbor and Roche Harbor. Based on the variance observed in our sediment samples from Friday Harbor and Fisherman Bay, we recommend a minimum of 20 grabs in each bay.

The next step is annual monitoring. Fixed sampling stations should be established in each bay for this purpose. Most laboratory analyses are within the capability of trained high school students. Supplies, such as solvents and antibodies, cost (at present) no more than \$25 per sediment sample tested.

If feasible, model indicator organisms should also be tested periodically to ensure good calibration of bioaccumulation estimates. Mussels (*Mytilus edulis*) are preferred in aquatic toxicology and occur naturally in all of the islands' working bays. Infaunal clams such as *Saxidomus giganteus*, the "butter clam" used in our Eastsound study (Barsh et al. 2009b), is also a reliable model, but longer-lived. A single pooled sample of >25 mussels or clams from each bay would suffice.

Above all, it is critical to integrate sediment monitoring into public considerations of island runoff and storm sewer management. Islanders are often critical and suspicious of government pronouncements regarding threats from runoff, and government should be in a position to meet their concerns with reliable, relevant local scientific data.

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1. Sediment grain size analysis, Fisherman Bay

Sample	Weight by grain size class, in grams						% Silts
	> 1 mm	0.5 mm	0.25 mm	0.1 mm	<0.1 mm	Total	
Grab 1	0.19	0.39	6.64	8.84	0.62	16.68	57
Grab 2	2.33	10.88	5.53	0.33	0.05	19.12	2
Grab 3	0	0.15	1.15	14.46	1.82	17.58	93
Grab 4	0.07	0.13	4.96	10.72	0.87	16.73	69
Core I top	0.05	0.04	0.55	12.81	1.96	15.41	96
Core I middle	0.05	0.25	1.46	11.73	2.19	15.68	89
Core I bottom	0.08	0.24	1.79	10.17	2.16	15.44	86
Core II top	0.08	0.06	1.47	11.22	2.02	14.85	89
Core II middle	0.11	0.21	1.52	9.39	4.23	15.46	88
Core II bottom	0	0.94	2.50	13.73	1.56	18.73	82
Core III top	0.02	0.01	1.17	11.93	1.62	14.75	98
Core III middle	0.07	0.10	1.21	10.83	3.66	15.87	91
Core III bottom	0.11	0.29	1.50	9.92	3.43	15.25	88

2. Sediment grain size analysis, Friday Harbor

Sample	Weight by grain size class, in grams						% Silts
	> 1 mm	0.5 mm	0.25 mm	0.1 mm	<0.1 mm	Total	
Core A top	0.41	0.61	2.76	16.3	6.83	26.91	86
Core A bottom	0.77	0.67	3.31	20.9	4.47	30.12	84
Core B top	0.24	1.01	15.0	13.9	4.97	35.12	54
Core B bottom	0.24	1.33	16.2	12.1	2.89	32.76	36
Grab C	0.18	0.60	2.19	20.5	3.32	26.79	89
Grab D	1.18	1.05	2.25	5.04	5.14	14.66	69

3. Sediment contaminants, Fisherman Bay

Sample	% Weight	Parts per million				% Silts
	TPH	Copper	Zinc	PAHs	Pyrethroids	
Grab 1	1.0	50	12	0.20	0.33	57
Grab 2	0.3	-	-	-	0	2
Grab 3	1.1	21	12	0.41	0.41	93
Grab 4	1.1	84	20	0.30	0.50	69
Core I top	1.4	89	22	0.25	0.35	96
Core I middle	1.7	32	15	0.03	0.16	89
Core I bottom	1.0	26	16	0.25	0.25	86
Core II top	1.6	70	25	0.42	0.33	89
Core II middle	1.4	27	12	0.17	0.01	88
Core II bottom	1.3	25	15	0.28	0.24	82
Core III top	1.6	81	30	0.49	0.33	98
Core III middle	1.4	20	18	0.16	0.24	91
Core III bottom	1.0	30	16	0.02	0.43	88
Means	1.2	46	18	0.25	0.30	

4. Sediment contaminants, Friday Harbor

Sample	% Weight	Parts per million			% Silts
	TPH	Copper	Zinc	PAHs	
Core A top	3.2	65	6.1	1.60	86
Core A bottom	4.8	35	6.3	0.80	84
Core B top	5.4	21	3.7	1.80	54
Core B bottom	2.8	36	4.8	0.34	36
Grab C	4.9	27	5.0	0.60	89
Grab D	0.6	-	-	1.25	69
Grab E	-	-	-	0.60	-
Grab F	-	-	-	0.42	-
Grab G	-	-	-	0.43	-
Grab H	-	-	-	0.28	-
Grab I	-	-	-	0.07	-
Means	3.6	37	5.2	0.79	

5. Trends over time in cores (% increase/decrease)

Sample	% Weight	Parts per million				% Silts
	TPH	Copper	Zinc	PAHs	Pyrethroids	
Fisherman Bay I	+40	+242	+38	NC	NC	+12
Fisherman Bay II	+23	+180	+67	NC	NC	+9
Fisherman Bay III	+60	+170	+88	+235	NC	+11
Friday Harbor A	-33	+46	-3	+100	-	+59
Friday Harbor B	+93	-42	-23	+429	-	+50