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Physico-chemical analysis of traditional vegetal salts obtained from three provinces of Papua New Guinea

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ABSTRACT

Objective: To determine the chemical constituents of the traditional vegetal salts and find out if they are safe to consume.

Methods: Seven different salts have been obtained from three provinces, of which five belong to Morobe Province. The cations were determined using inductively coupled plasma atomic emission spectroscopy and anions using titrimetry, gravimetry and spectrophotometry. Others like solubility, electrical conductivity, pH, antimicrobial, Fourier transform infrared spectral and volatility studies have been carried out for these salts.

Results: While few salts were found to be stable, others were deliquescent; and the colour varies from white to black through yellow and brown. It was found that the potassium ion was dominant while others including sodium and calcium were found in lower concentrations. For the first time, certain *d*-block metal concentrations were measured though most of them were found to be present at very low levels. Other parameters like volatility, solubility, electrical conductivity, antimicrobial and Fourier transform infrared spectral studies were carried out for the first time for these vegetal salts in Papua New Guinea.

Conclusions: The salt's deliquescence could be correlated to the presence of anions like carbonate, bicarbonate and hydroxide which strongly contribute towards it. Similarly, solubility and conductivity of the salts could be correlated well. The salts were found to be harmless for consumption, but for the high potassium content.

1. Introduction

Salt as a very useful trade commodity has been the subject of interest since its discovery and inception to humans. Adequate salt amounts are very much in need for fulfilling several basic requirements such as balancing nutritional diets, enhancing taste, preserving foods and feeding livestock, to name a few. Its disinfectant and hair-removing properties further enhance the wholesome utility in medical and industrial applications as well[1-3]. In ancient times, salt was considered as a very precious commodity and was often used as a currency to exchange for supplies like pigs, woven bags, cowries, tools, weapons like steel axes, machetes, ritual

objects, to name a few[4-7]. Majority of the salt is produced near the coastal areas, and people faraway from these areas are deprived of this basic commodity. Such crises, led to the onset of large-scale salt production as well as transportation to other locations[3].

Five major sources have been found[1,3,4,8] to be useful for producing salt: (1) evaporating the sea water using "salt-making potteries"; (2) evaporating water from the salt springs; (3) soaking the dried grass in saline water collected from salt springs followed by burning the grass in firewood and evaporating the filtrate; (4) burning salt-containing grass or plants, lixiviating the ash and evaporating the solution; and (5) mining the salt rocks from caves and ground to get the "geological salt". Among them, vegetal salts deserve special mention due to their ease of preparation and wide spread utility. Burning grasses, ferns, herbs, shrubs, palms, leaves, barks, shoots, stems or even the whole plant and tree are the major adapted protocols[9]. Throughout the world more or less the same methodology has been practiced, but with little variations in the

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plants, tools and equipment utilized. In general, plant materials will be burnt in selected firewood, the ashes will be lixiviated through the filters and the resulting brine will be slowly dehydrated to obtain the dry salt[3,10-15].

1.1. Salt and ash production in Papua New Guinea (PNG)

Salts and salt ashes in the PNG area were primarily used in initiation ceremonies and inter-tribal exchanges, rather than for culinary aspects[13,14]. In some regions, direct usage of black ash without any refinement was also noticed. The Gidra-speaking Papuans of the Oriomo Plateau, lowland of PNG, consume the ashes of the tree species *Melaleuca leucadendron* and *Acacia mangium*. These ashes contain relatively higher levels of sodium (100-200 mg)[16]. However, they are alkaline and hence are not palatable as well as are injurious to health[17]. Ashes from the base leaves of midrib from sago palm (*Metroxylon* sp.) were also used as salt substitute by the Saniyo-Hiyewe community from the East Sepik Province. It contains around 0.59% Na⁺, 26.3% K⁺, 11.7% Ca²⁺ and 3.4% Mg²⁺[9]. The Baruya speaking sector of Eastern Highlands Province (EHP) exhibited a specialized art of making salt[18,19]. The production techniques, chemical composition, appearance, and taste as well as the uses were different from those made by the Angans that were exclusively used for the *Coix gigantea*[14]. They settled in places where salt-containing springs were in abundance, and salt has been stored as bars. The ash salt from the fronds of epiphytic fern, *Asplenium* sp. was mixed with water and drunk as medicine too[20,21]. The salt tasting plant species *Saccharum klossii* were used as preservatives by the Marawaka people (EHP) for masking the bitter tastes of herbal preparations[22].

Tribes own the land in Oceania area, especially in Melanesia than the government[23]. Since the people lived inland, salt trade did not reach those areas and so burning of plants was the only means of getting salt into their diets. Overall, ethnic groups in different provinces have various ways of producing vegetal salts from plant species. In order to take advantage of the health benefits of vegetal salts, an in-depth chemical analysis of the inorganic constituents has to be carryout. Towards this end, herein physicochemical analyses of seven vegetal salts selected from three provinces (Morobe, Chimbu and East Sepik) have been reported.

1.2. General preparation of traditional vegetal salts in PNG

Most of the plants used for salt production are usually found near river streams, swampy and marshy lands and are cultivated by the people living in the area. When matured, they are harvested, gathered, heaped up and sun dried for ~10 h. A specially designed hut is erected with an inclined roof and open sides to withstand heavy smoke and flames and for good ventilation; the dried material is burnt in the fire. About 5-6 tons of the plants will be burnt to produce ~5 kg of salt. Such salts were produced by certain techniques using specialized indigenous equipments like pots, ladles, pipes, filters and containers. Each clan has its own procedure and is

usually considered as the proprietary. The Anga clan of Marawaka (EHP) produce their “Anga salt” in a more classical way of crafted artwork. A nice display of their equipments (Figure 1) can be found in J.K. McCathy Museum-Archaic Technology, Goroka, EHP.

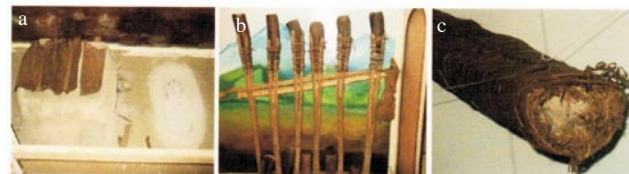


Figure 1. Anga salt making process: the salt extract evaporated using an oven called “sai-sito” (a); bamboo tubes called “iraigeye” that pass the filtrate out (b); and salt cake called “livadagala” wrapped up in dried banana leaves ready for trade (c).

These practices were inherited from their ancestors and hence considered as confidential intellectual property for the benefits of their immediate and extended families. Each clan uses particular plant types only and usually one type of plant.

2. Materials and methods

All the reagents used throughout the study were of analytical grade. Deionised water [Merit-W4000 (UK) water still] was used. pH and conductivity studies were carried out on 1% salt solutions after filtering the residues, using TPS Labchem-pH meter and TPS Labchem-conductivity meter at (25 ± 0.3) °C and 24.1 °C, respectively. Inductively coupled plasma atomic emission spectroscopic analyses were carried out at National Analytical and Testing Services Limited, PNG University of Technology (PNGUT), Lae, PNG for determining the amounts of cations, SiO₂ and P_{total}. The anions chloride (Mohr titration), carbonate, bicarbonate, hydroxide (acid-base titrations) and sulphate (gravimetry) were determined using the reported procedures[24]. The samples were mixed with KBr to prepare pellets and Fourier transform infrared (FTIR) spectra were recorded using JASCO FT/IR-4100 spectrophotometer. The botanical names of the plant species were identified by Herbarium Division, Department of Forestry, PNGUT.

2.1. Moisture content, volatile content and LOI studies

Moisture and volatile contents and loss on ignition (LOI) were determined using thermal oven (Binder, Australia) and muffle furnace (S.E.M., Isai Pty. Ltd., Australia). Around 1.0 g of the salt was placed in a dry crucible with lid and heated in a thermal oven at 105 °C for 2 h, later cooled and weight was measured. It was then heated in a muffle furnace at 350 °C for 2 h, cooled and the loss in weight was estimated. Subsequently, it heated in a muffle furnace at 650 °C for 2 h, cooled and the loss in weight was estimated[25].

2.2. Antimicrobial studies

Four human pathogens commonly found and encountered in PNG were selected: *Bacillus cereus* (*B. cereus*) and *Staphylococcus aureus* (Gram-positive) with incubation temperatures of 30 °C and

37 °C respectively; *Escherischia coli* and *Klebsiella pneumonia* (Gram-negative) with incubation temperature of 37 °C. One percent of salt solutions were prepared and a control sample with the same concentration and volume was prepared using common salt (NaCl) for comparison.

Preparation of sterile disc[26]: Satorius filter pad (45 mm) was punched into 6 mm discs and sterilized. Each disc was impregnated with 30 µL of the salt solution using micropipette. Precautions were taken to prevent the flow of the solution from the discs to the outer surface. The disc was then allowed to air dry at room temperature.

Assay of antimicrobial activity using disc diffusion method[26]: Using a micropipette, 100 µL of the test organism was transferred on to sterile nutrient agar plate and spread throughout the agar surface using glass spreader. The disc was then placed in the agar surface using sterile forceps and the plates incubated at 37 °C for 24 h. After incubation, the diameter of zone of inhibition formed around each disc was measured in millimetres.

3. Results

Table 1 provides information on the plant species and parts used for the traditional salt production in the three provinces (Morobe, Chimbu and East Sepik). Details on seven different types of salts collected from these three provinces are listed in Table 2. Aseki and Sepik River powder salts are found to be quite deliquescent when they are exposed to open atmosphere. All of them are microcrystalline except the Menyamya block salt that was found to be very hard and requires big size mortar and pestle to break. On the other hand, the Yamap powder salt was amorphous.

Table 1

Plants used in the present day for salt making in Chimbu, Morobe and East Sepik Provinces.

Place (Province)	Plant species	Family	Parts used	Vernacular name
Kup (Chimbu)	<i>Coix lacryma-jobi</i> L.	Poaceae	Whole plant	Engre muno
	Unknown sp.		Whole plant	Yawak
Hekwangi (Morobe)	<i>Impatiens hawker</i>	Balsaminaceae	Whole plant	Wakuye
	<i>Polytoxa macrophylla</i>	Poaceae	Whole plant	Hyaljaale
	<i>Coix lacryma-jobi</i> L.	Poaceae	Whole plant	Ee-wye
Oiwa, Aseki (Morobe)	<i>Coix gigantea</i>	Poaceae	Leaves, stems, branches	Mun-haka
	<i>Ficus elastic</i>	Moraceae	Leaves, stems, branches	Yangwa
	<i>Barringtonia calyptocalyx</i>	Lecythidaceae	Leaves, stems, branches	Hamewa
	<i>Dicksonia</i> sp.	Cyatheaceae	Leaves, stems, branches	Wombiyoka
	<i>Pandanus</i> sp.	Pandanaceae	Nuts along with shells	Hakuwa
	<i>Piper</i> sp.	Piperaceae	Leaves, stems, branches	Kungha
Yamap (Morobe)	<i>Ficus</i> sp.	Moraceae	Barks only	--
	<i>Asplenium nidus</i>	Aspleniaceae	Whole plant	--
	<i>Barringtonia calyptocaly</i>	Lecythidaceae	Leaves and barks	--
Kubkain (East Sepik)	<i>Musa</i> sp.	Musaceae	Peels only	--
	<i>Ipomoea batatas</i>	Convolvulaceae	Peels only	--
	<i>Colocasia esculenta</i>	Araceae	Peels only	--

Table 2

Details on various salt samples collected from Chimbu, Morobe and East Sepik Provinces of PNG.

Salt	Place	District	Province	Latitude/Longitude	Ethnic group	Salt colour	Salt texture	Other details
Kerowagi powder	Kup	Kerowagi	Chimbu	5° 58' S / 144° 48' E	Siku	Black	Microcrystalline	Stable to open air
Menyamya block	Hekwangi	Menyamya	Morobe	7° 10' S / 145° 55' E	Menye	Greyish white	Hard block	Stable to open air
Menyamya powder	Hekwangi	Menyamya	Morobe	7° 10' S / 145° 55' E	Menye	Cream yellow	Microcrystalline	Stable to open air
Aseki powder (sample 1)	Aseki	Menyamya	Morobe	7° 22' S / 146° 16' E	Kukuku pate	Light brown	Microcrystalline	Deliquescent when kept open
Aseki powder (sample 2)	Aseki	Menyamya	Morobe	7° 22' S / 146° 16' E	Kukuku pate	Dark brown	Microcrystalline	Deliquescent when kept open
Yamap powder	Yamap	Huon Gulf	Morobe	7° 4' S / 146° 39' E	Mumeng	Cream yellow	More amorphous	Stable to open air
Sepik River powder	Kubkain	Ambunti	East Sepik	4° 18' S / 142° 19' E	Sanyo-Hiyewe	Light brown	Microcrystalline	Deliquescent when kept open

Table 3 narrates the comparison of the current results with the published data for the cations and anions. some of the s-, d- and p-block elements (Li, Na, Mg, Ca, Sr, Ba, V, Mo, Mn, Fe, Cu, Zn and B) are present in low concentrations (<0.001%-1.341%) while potassium is the most common and present in abundance (21.64%-45.02%) in the salt samples of PNG. The range of pH observed is 9.2-10.1 for the seven types of salts. Aseki (sample 1) and (sample 2) and Sepik River salts contain higher amounts of anions that contribute to deliquescence, viz., 19.52%, 10.65% and 11.23% respectively. The Kerowagi salt obtained from Chimbu Province stood high in terms of insolubles (34.45%) which was dark black in colour and resembled charcoal, while Menyamya powder and Aseki salts had low amounts of insolubles 0.52% and 0.60%, respectively. Menyamya salts displayed values (14 165 and 14575) closer to NaCl solution thus indicating higher amounts of chloride (17.00% and 25.06% respectively). FTIR spectra and data for the six salts except Aseki (sample 1) are provided in Figure 2 and Table 4 respectively. Table 5 shows the results obtained from volatility studies and the ash content denotes the amount of minerals available in each of the salts.

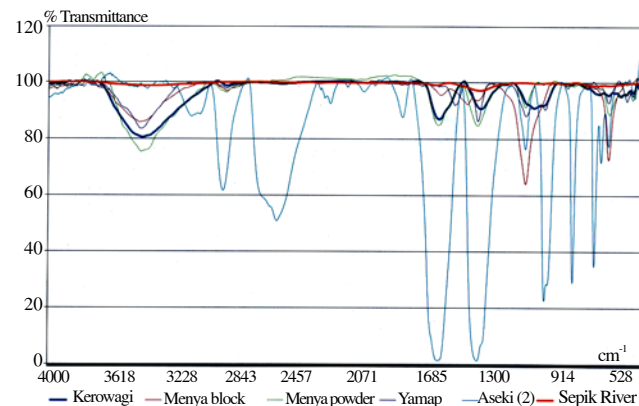


Figure 2. FTIR spectral comparison of various vegetal salts of PNG.

Table 4

FTIR spectral data for the traditional salts of PNG.

Salt	IR (cm ⁻¹)
Kerowagi powder	1 636 (vs), 1 384 (vs), 1 065 (vs, br)
Menyamya block	1 633 (vw), 1 472 (w), 1 410 (w), 1 106 (vs), 635 (vs)
Menyamya powder	1 644 (vs), 1 410 (vs), 1 097 (s), 636 (s)
Aseki (sample 2)	2 908 (s), 2 597 (vs, br), 1 814 (w), 1 644 (vs), 1 410 (vs), 1 118 (w), 1 011 (vs), 829 (s), 700 (s), 657 (m), 614 (w)
Yamap powder	1 515 (vs), 1 365 (vs), 1 086 (vs), 979 (s), 614 (s)
Sepik River	1 666 (m), 1 386 (vs), 851 (w), 679 (m), 593 (m)

s: Strong; vs: Very strong; m: Medium; w: Weak; vw: Very weak; br: Broad.

Table 3

Comparison of cations, anions and other properties of traditional vegetal salts.

Type	Location in Periodic table	Ions	Yamap	Aseki (1)	Aseki (2)	Menya Block	Menya powder	Kerowagi	Sepik River	1*	2*	3*	4*	5*	6*	7*	Longi	Jiwika		
Cation (wt. %)	s-Block	Li ⁺	n.d.	n.d.	n.d.	<0.001	<0.001	n.d.	n.d.	0.130	<0.100	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.		
		Na ⁺	0.070	<0.001	0.081	0.150	0.100	0.030	0.396	0.200	0.140	0.410	20.100	0.040	0.010	0.050	0.370	1.500		
		K ⁺	31.650	45.020	41.590	29.240	28.770	21.640	35.680	29.600	27.100	38.100	10.600	44.640	36.060	28.900	<0.010	0.020		
		Mg ²⁺	0.270	0.042	0.231	0.290	0.020	0.660	0.222	n.d.	n.d.	0.030	0.300	0.000	0.000	<0.010	<0.010	0.030		
		Ca ²⁺	1.250	0.030	1.341	0.310	0.320	0.680	1.110	12.600	17.800	2.370	0.800	0.000	0.000	0.140	0.020	0.120		
		Sr ²⁺	<0.001	<0.001	<0.001	0.004	<0.001	0.005	<0.001	n.d.	n.d.	n.d.	n.d.	0.030	0.000	0.010	<0.010	0.030		
		Ba ²⁺	<0.001	<0.001	<0.001	0.002	<0.001	0.006	<0.001	n.d.	n.d.	n.d.	n.d.	0.020	<0.010	0.100	n.d.	n.d.		
		p-Block	B	0.004	0.005	0.010	0.005	<0.001	0.006	0.003	n.d.	n.d.	n.d.	n.d.	<0.010	<0.010	0.020	n.d.	n.d.	
			d-Block	V	<0.001	<0.001	<0.001	<0.001	<0.001	<0.001	<0.001	n.d.	n.d.	n.d.	n.d.	0.070	<0.010	<0.010	n.d.	n.d.
				Mo	<0.001	<0.001	<0.001	<0.001	<0.001	<0.001	0.001	n.d.	n.d.	n.d.	n.d.	<0.010	<0.010	<0.010	n.d.	n.d.
	Mn			<0.001	0.001	<0.001	0.006	<0.001	0.018	<0.001	n.d.	n.d.	n.d.	n.d.	<0.010	0.000	<0.010	n.d.	n.d.	
	Fe			<0.001	<0.001	0.009	<0.001	0.020	0.151	<0.001	n.d.	n.d.	n.d.	n.d.	0.000	0.000	0.000	n.d.	n.d.	
	Cu			<0.001	0.002	<0.001	<0.001	<0.001	0.014	0.003	n.d.	n.d.	n.d.	n.d.	0.000	<0.010	<0.010	n.d.	n.d.	
	Zn			<0.001	<0.001	<0.001	<0.001	0.040	0.018	<0.001	n.d.	n.d.	n.d.	n.d.	<0.010	0.000	0.000	n.d.	n.d.	
	Anion (wt. %)	Cl ⁻		16.670	4.880	3.090	25.060	17.000	6.160	3.300	25.300	18.900	22.100	26.300	26.120	2.810	15.260	0.400	2.240	
		SO ₄ ²⁻	11.090	2.580	4.710	6.390	3.820	15.910	13.880	<1.000	<1.000	13.700	n.d.	14.870	19.620	55.900	0.000	0.010		
		CO ₃ ²⁻	2.940	3.210	9.510	0.000	0.000	2.470	4.990	n.d.	n.d.	1.540	n.d.	0.910	17.960	9.010	n.d.	n.d.		
HCO ₃ ⁻		1.420	13.030	0.000	0.000	0.000	3.540	4.640	n.d.	n.d.	3.730	n.d.	n.d.	n.d.	n.d.	0.060	0.030			
OH ⁻		0.870	3.280	1.140	0.000	0.000	0.190	1.600	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.			
SiO ₂ [#]		0.010	0.010	0.010	<0.001	<0.001	0.020	0.010	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.			
PO ₄ ^{3-#}		<0.001	<0.001	0.250	<0.001	<0.001	<0.001	<0.001	n.d.	n.d.	n.d.	n.d.	0.030	0.020	0.010	n.d.	n.d.			
pH			9.2	9.3	9.2	9.9	10.1	10.1	9.9	n.d.	n.d.	n.d.	n.d.	n.d.	8-14		8.5	7.1		
Conductivity (µS/cm)			13485	11095	10415	14165	14575	6575	12735	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	15900	64000		
Anions contributing to deliquescence [CO ₃ ²⁻ , HCO ₃ ⁻ , OH ⁻]			5.23	19.52	10.65	0.00	0.00	6.20	11.23	--	--	--	--	--	--	--	--	--		
Insolubles (%)		8.15	0.60	6.00	9.5	0.52	34.45	6.09	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.			
Total minerals (%)		66.24	72.09	61.72	61.70	50.09	51.52	65.83	67.83	63.95	81.98	58.1	86.73	76.48	109.4?	0.86	4.00			
Balance ionic (%)		33.76	27.91	38.28	38.30	49.91	48.48	34.17	32.17	36.05	18.02	41.9	13.27	23.52	--	99.14	96.00			
KCl (wt. %)		48.32	49.90	44.68	54.90	46.00	27.80	38.98	54.9	46.00	60.2	57.00	70.76	38.87	44.16	--	--			
NaCl (wt. %)		--	--	--	--	--	--	--	--	--	--	--	--	--	--	--	0.77	3.74		
Reference		Present study								[11]	[11]	[27]	[27]	[12]	[12]	[12]	[30]	[30]		

1*: *Asplenium acrobryum*; 2*: *Asplenium nidus*; 3*: *Coix gigantea*; 4*: *Eriocoulon australe*; 5*: *Thurnia sphaerocephala*; 6*: *Ecclinusa bullata*; 7*: *Gustavia hexapetala*; n.d.: Not determined; #: Determined using inductively coupled plasma atomic emission spectroscopy (ICP-AES) as P_{total}; ^: The plant ash was completely digested with concentrated HNO₃ and elements were determined using flame photometry; the reported values get contributions from both soluble as well as insoluble portions; ® SiO₂: Probably present as the respective oxoanion (viz., silicate) because of the prevailing alkaline conditions which are suitable for the formation of silicates; 1*-4* from PNG; 5*-7* from Columbian and Peruvian Amazon; Longi and Jiwika from Papua Province, Indonesia.

Table 5

Loss in weight reported from volatility studies (%).

Salt	Moisture content	Volatility content	LOI	Total volatiles	Ash (mineral) content	Colour of the final ash	Ash when exposed to open air
Kerowagi	31.37	17.24	8.12	56.72	43.28	Dull white	Sticky
Aseki (1)	4.23	19.74	1.24	25.22	74.78	Dull white	Turned to liquid
Aseki (2)	7.13	22.12	1.21	30.46	69.54	Dull white	Turned to liquid

The effect of salt solutions on *B. cereus* was shown in Figure 3. All the seven salt solutions showed no inhibition zone against *B. cereus*.

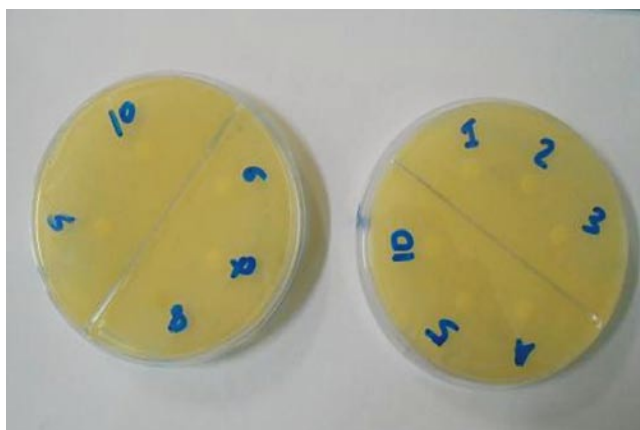


Figure 3. Plates showing the action of the seven salt solutions on *B. cereus*.

1: Kerowagi powder; 2 & 3: Menyamya block salt; 4 & 5: Menyamya powder salt; 6: Aseki powder (sample 1); 7: Aseki powder (sample 2); 8: Yamap powder; 9: Sepik River powder; 10: Common salt (NaCl) which is the control sample.

Figure 4 shows possible exchange pathways of soluble minerals between water, soil and plant. While the plant and soil are in static states, the water body is always in a state of dynamism.

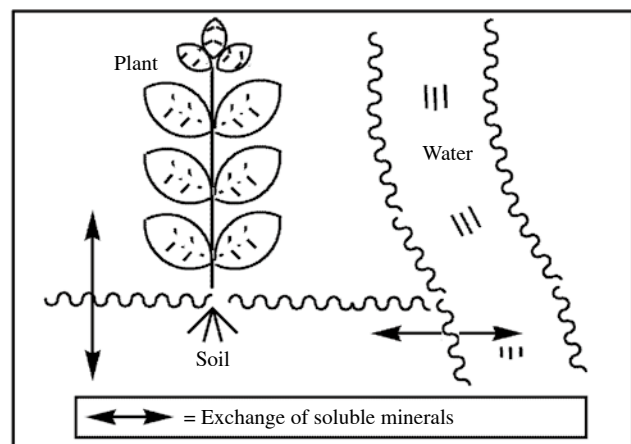


Figure 4. Possible exchange pathways for minerals among plants, water and soil bodies.

Table 6

Comparison of the minerals content on the basis of average daily intake (~10 g) and the RDI (mg/d).

Salt	Li ⁺	Na ⁺	K ⁺	Mg ²⁺	Ca ²⁺	Sr ²⁺	Ba ²⁺	B	V	Mo	Mn	Fe	Cu	Zn
Yamap	n.d. [#]	7.0	3165	27.0	125.0	<0.1	<0.1	0.4	<0.1	<0.100	<0.1	<0.1	<0.1	<0.1
Aseki (1)	n.d. [#]	<0.1	4502	4.2	3.0	<0.1	<0.1	0.5	<0.1	<0.100	0.1	<0.1	0.2	<0.1
Aseki (2)	n.d. [#]	<0.1	4159	23.1	134.1	<0.1	<0.1	1.0	<0.1	<0.100	<0.1	0.9	<0.1	<0.1
Menya block	<0.1	15.0	2924	29.0	31.0	0.4	0.2	0.5	<0.1	<0.100	0.6	<0.1	<0.1	<0.1
Menya powder	<0.1	10.0	2877	2.0	32.0	<0.1	<0.1	<0.1	<0.1	<0.100	<0.1	2.0	<0.1	4.0
Kerowagi	n.d. [#]	3.0	2164	66.0	68.0	0.5	0.6	0.6	<0.1	<0.100	1.8	15.1	1.4	1.8
Sepik River	n.d. [#]	39.6	3568	22.2	111.0	<0.1	<0.1	0.3	<0.1	0.100	<0.1	<0.1	0.3	<0.1
RDI	n.s. [#]	2400.0	3500.0	350.0	1000.0	5.0	14.0	<20.0	<1.8	0.075	5.0	15.0	2.0	15.0

n.d.[#]: Not determined; n.s.: Not specified.

The minerals content for all the salts were calculated and compared with the recommended daily intake (RDI) values as shown in Table 6. The potassium content in Aseki and Sepik river salts exceeded the RDI value. Iron content in Kerowagi salt was higher than the RDI value.

4. Discussion

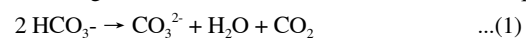
It is clear from the present study that several new plant species, which are very different from the published reports[11,14,16,27] are used for the salt production. As many as fourteen different ethnic groups have been reported to be actively involved in salt preparations[9,14]. Some of these places have been visited by the author to collect salt and plant samples, and to interact with the local people who geographically do not belong to the coastal areas, but are not very interior as compared to other provinces and ethnic groups of PNG. They prepare salts by burning the plants, but it is not surprising to note that seawater was not used though their proximity to the sea-coast is close because of occasional restriction in their movements due to inter-tribal or local disputes.

4.1. Cationic and anionic constituents

The individual insoluble residues are not analysed as the salts are consumed either by sprinkling over food or adding during cooking; however, both soluble and insoluble portions were digested using a minimum *aqua regia* (HNO₃:HCl, 1:3, 4 mL) and the cations thus present are determined using ICP-AES. Based on the limited reports available[11,12,14,16,27], common cations present in various salt samples are Na, K, Mg and Ca (macronutrients), and Li, Sr, Ba, B, V, Mn, Cu, Zn and Mo (micronutrients). With rich abundance of minerals in the Earth's crust of PNG, it is not surprising to note their presence in the salt samples. Most of the elements (especially the *d*-block, *viz.*, V, Mo, Mn, Fe, Cu and Zn and the *p*-block, *viz.*, B and some of the alkali and alkaline earth metals) were determined for the first time for the authentic traditional vegetal salts of PNG. Literature reports are available only for the analysis of the plant ashes obtained by burning the plants under laboratory conditions[11]. Majority of the elements were not considered for analysis by the earlier researchers. Though the results in most of the cases show <0.01%, it is now clear that these elements are not present in the

salts. Lithium was determined only for Menyama block and powder salts and found to be <0.001%. Hence, lithium was not considered for the other five salts. The literature data provided for the salts are old and either flame photometry or atomic absorption spectrometry was used for the determinations. But, in the present work, the salts freshly prepared by those ethnic people presently living in those villages were subjected to ICP-AES analysis and thus renewed the data with the advent of additional sophistications available with such techniques and instrumentations. Some of the rare cations were analysed because: (a) their ionic radii have similarities with some of the common elements, and hence chances are there to be preferentially absorbed by the plants; and (b) rich mining activities are going on in and around PNG. None of the salts analysed in the past or in the present study revealed the presence of nitrate ion which is quite unexpected because of their more thermal lability[28], unlike reported for Kurima salt of Indonesia[29,30]. The % values for the cations and anions are found to be different from the reported ones and could be attributed to: (a) possible combination of different plant species for producing salt; (b) contributions from both water-soluble and insoluble portions of the plant ash which was completely digested with acid and then metal analysis carried out[11]; and (c) the use of better and advanced analytical methods now compared to the past which were determined some 25-30 years ago.

The common and usual anions that are present are chloride, sulphate, some thermally labile anions like carbonate and bicarbonate, and rarely phosphate. However, the presence of thermally labile anions and their amount largely depends on the pyrolytic conditions used to prepare the salts because they can decompose according to the equations 1-3. The presence of bicarbonate, carbonate, oxide and hydroxide ions makes the brine solution alkaline because solutions of alkali and alkaline earth chlorides and sulphates are expected to be neutral with a pH of 7. In fact, sulphates are also capable of decomposing during the ashing process[31] as shown in equation 4. This also partly explains the deliquescent nature of some salts. In line with observations made in this study, the Aseki (sample 1) and (sample 2) and Sepik River salts contain higher amounts of anions that contribute to deliquescence.





From this study, it is very clear that there is no order or sequence in distribution of both cationic and anionic species because of wide ranges observed between plants. While lithium was only observed by Croft and Leach[11], other researchers could not detect it or did not determine it. Based on the given data, it is clear that the traditional vegetal salts cannot act as salt substitutes because of very low sodium content. A study conducted by Dickie and Malcolm[17] on rats indicated that the salts, when used in suitable quantity contain enough NaCl for the physiological needs in higher animals and humans in spite of containing large amounts of potassium ions. Prolonged studies[32,33] suggested that the body retained sodium in spite of large intake of potassium (through the traditional salts). Majority of the cations have a specific role to play in the biological systems[34,35]; while some are beneficiary, others exert a high level of toxicity in the human metabolism. Also, those anions which combine with certain cations to produce less soluble salts like CaSO_4 , $\text{Ca}_3(\text{PO}_4)_2$, to name a few, retained along with the residual ash while filtration process was carried out.

4.2. Role of anions in enhancing the taste of the salts

Cations play a minor role in determining the salt taste while anions have major influence[12,14]. Salts with a high concentration of chloride and carbonate are perceived as “sweet and cool” and “strong and hot”, respectively by the Witoto Indians. On the other hand, the taste produced by sulphate combines with the sensation by the other two anions to give “biting and insipid”. Since those of Witoto salts contain very low sodium content (0.007%-0.391%), the real “salty” taste could not be experienced as saltiness arises only when sodium cations are combined with chloride anions. In the present study, these three taste deciding anions (chloride, carbonate and sulphate) are present in larger amounts (17.31%-31.45%) for all the salts except for Aseki (sample 1) salt (10.67%).

4.3. Total mineral content

The total mineral content reported by Weller *et al.*[29] for the two salts used in Indonesia was very low (0.86% and 4.00%); probably the authors would have overlooked in converting the readings from mg/L to mg/kg after incorporating the concentration of the solution taken for the analysis. On the other hand, those reported by Croft and Leach[11] were found to be in the acceptable range (58.1%-81.98%); the difference being Weller *et al.* collected the vegetal salts from various groups, while Croft and Leach prepared them in the laboratory from plant species. In the present study, the values were found to be in between 50.09%-72.09%. There are several reports that highlight[12] the burning of the stem part of *Gustavia hexapetala* to ash for extracting the salt. However, the total mineral content reported, 109.4%, is not acceptable suggesting an oversight in the estimation of cations and/or anions.

4.4. Solubility studies

This study indicated a wide range of solubility with varying degrees of insolubles. In most cases, the colour of the insoluble residue was found to be gray, pale-brown or black. The Kerowagi salt obtained from Chimbu Province stood high in terms of insolubles (34.45%) which was dark black in colour and resembled charcoal, while Menyamya powder and Aseki salts had low amounts of insolubles 0.52% and 0.60%, respectively. This observation corroborates well with the published report[36], according to which huge amounts of wood were burned every year for extracting the salt and the charcoal formed during the process were transmitted downhill. When the charcoal pile was washed down, the salts picked up charcoal and wood ash to an extent of 10%-50%. The solubility test was mainly done to identify the extent of insoluble material present as their presence will have impact on the overall conductivity.

4.5. Electrical conductivity studies

Previous studies have not tested for electrical conductivity. This is done here for the first time. The electrical conductivity was determined from 1% solutions at 24.1 °C. The results indicate the amount of electrolytes (dissolved ionic constituents) present in the solution, which in-turn suggests the amount of the salt. The solubilities of the salts varied drastically and hence the conductivity values. The comparison of seven salts indicated a maximum conductivity (14575 $\mu\text{S}/\text{cm}$) for the Menyamya powder salt (which has minimum insolubles, 0.54%) and minimum (6575 $\mu\text{S}/\text{cm}$) for the Kerowagi black salt (which has maximum insolubles, 34.45%). For a better comparison, a blank run was made for the distilled water used for preparing the salt solutions, and its conductivity, 5.06 $\mu\text{S}/\text{cm}$, was subtracted from the measured values, while a 1% NaCl solution showed 16860 $\mu\text{S}/\text{cm}$. Since temperature of the measurement was kept constant, the electrical conductivity depends on three factors: (i) percent solubility of salts; (ii) mobility of ions, which is dependent on the size of the ions (Cl^- is smaller in size compared to SO_4^{2-} , for example); and (iii) valency of the ions (Na^+ , Ca^{2+} , Fe^{3+} , for example). The combination of ions as well as their presence at various proportions changes the conductivity values. The salts in the present study have comparable values with the Longi and Kurima salts of Papua Province, Indonesia, while Jiwika salt of Papua Province, Indonesia was found to have very large conductivity values[29]. It appears that in Jiwika salt, the ions that are present in the solution act as strong electrolytes and thus are able to conduct more electricity than the other salts.

4.6. pH studies

The pH of the salts was found to lie in the narrow alkaline range of 9.2-10.1. This observation clearly supports the presence of alkaline anions like hydroxide, carbonate, *etc.* The pH of Kurima (Papua

Province, Indonesia) salts was found[29] to be close to the neutral range (7.1-8.4), which do not possess any such alkaline anions.

4.7. Antimicrobial studies

This is the first time that such a study has been carried out because the salts were usually prepared in an unhygienic way in the open area near the house. For all the cases including the common salt, no inhibition zone could be identified and the effect seemed to be more or less the same for all the salts. Either the chosen concentration (1%) was insufficient or the chosen bacteria were not responding to the salts.

4.8. FTIR spectral studies

FTIR studies haven't been reported for any of the traditional salts prepared either in PNG or elsewhere. In all the cases a very broad and typical band was observed in the region $3680-2840\text{ cm}^{-1}$ that could be attributed to the presence of moisture and its ingress into the vegetal salts[37]. The polyatomic anions present in the salts could be identified from the characteristic bands: carbonate (CO_3^{2-}) $1450-1410\text{ (vs)}$ and $880-860\text{ (m)}\text{ cm}^{-1}$; sulphate (SO_4^{2-}) $1130-1080\text{ (vs)}$ and $680-610\text{ (m, w)}\text{ cm}^{-1}$; and phosphate (PO_4^{3-}) $1100-1000\text{ (vs, br)}\text{ cm}^{-1}$ [37]. In the present study, since the salts contain different polyatomic anions in different proportions, there could be some common overlapping bands and could obscure some vital information. Also, most of the bands are found to be occurring in all the six salts [except Aseki (sample 1)], thus suggesting the presence of common anions.

4.9. Volatility studies

The volatility data is reported for Aseki and Kerowagi salts only as other traditional salts were purely inorganic in nature, the salts from Aseki contained organic compounds and Kerowagi salt is black in colour suggesting the presence of charcoal (carbon). During the Aseki salt preparation, a tree sap was added which obviously contains organic compounds. The loss in weight reported from the volatility studies narrates the obtained results as well as the ash content that denotes the amount of minerals available in each salt. While the % of insolubles in Kerowagi salt is only 34.45%, the total volatiles % are found to be 56.72% indicating that certain species have probably volatilized or decomposed during the process. The Kerowagi salt was found to be quite stable in the open air and its moisture content was found to be extremely high (31.37%). After the moisture tests, all the three salts caked out thus indicating the presence of significant amounts of moisture in them. After the volatility tests, they appeared gray in colour thus indicating that the colour-contributing substances in them were volatilized at the test temperature ($650\text{ }^\circ\text{C}$). After the LOI tests, they appeared dull white. Based on the different values obtained for both Aseki salts (samples 1 and 2), it could be concluded that their preparation methods are different either in terms of the type of plants used, type of milk sap used or the processing technique.

From the solubility and volatility studies, it became clear that carbon particles generated during the plant burning are also being consumed by the people along with the Kerowagi salt. Whether such carbon particles are toxic or not is still a topic for debate. However, recent epidemiological studies indicated that the combustion-generated fine carbon particles are directly associated with lung cancer and heart diseases including asthma[38], while the other school of thought says that they have extremely low toxicities to humans and are resistant to dissolution or chemical attack even in the acidic pH range especially in the digestive tracts; but once entered into the body tissues, it remains there for an prolonged periods of time[39], and could cause other complications. After the LOI tests, when the three residual ashes were exposed to open atmosphere, they became deliquescent indicating the presence of mostly oxides and hydroxides thus supporting equations 1-4. The high temperature employed in the LOI test ($650\text{ }^\circ\text{C}$) is sufficient enough to cause decompositions to the anions like bicarbonate, carbonate and to some extent sulphate[31].

4.10. Indigenous view on salts bearing medicinal values

Of the seven salts, Menyamya and Aseki salts were used as medicines. Probably, when they are consumed, the required inorganic mineral nutrients (both micro- and macro-) are being delivered to the body and hence the people feel fresh and rejuvenated. Though they are of plant origin, the organic parts of the plants, which probably might have some medicinal values, are burnt completely to ash from which the inorganic salts are obtained. So, the presence of simple cations and anions would not have contributed to the medicinal properties, as claimed by the local people, and it is the opinion of this author that it could be an indigenous or a cultural view.

4.11. Why only certain plants and why not others?

Based on the data collection and careful analyses, it is really very puzzling to note that only selected plants were used for making the vegetal salts by traditional methods whereas all other plant species grown around the banks of river streams or salt springs, and common garden backyards should also give more or less the same distribution of cations and anions as this study. From the present study, it is very clear that some of the *s*-, *d*- and *p*-block elements (Li, Na, Mg, Ca, Sr, Ba, V, Mo, Mn, Fe, Cu, Zn and B) are present in low concentrations ($<0.001\%$ - 1.341%) while potassium is the most common and present in abundance (21.64% - 45.02%). The same was also reported[12] and the values are $<0.001\%$ - 0.630% and 27.00% - 48.50% respectively for almost all the salts obtained from various plant species belonging to different families.

4.12. Why only plants and why not sea/spring waters?

A cautious analysis of those plant species revealed some interesting facts and questions that remain unanswered. The choice of using these

plants probably lies in the fact that they grow in and around water bodies (marshy, swampy lands and river banks) and as expected they have adapted to absorb plenty of water which became very clear on proper examination of one of the fresh plant species obtained from Menyamya. Therefore, they naturally contain more of soluble inorganic salts, and such of those species were deliberately selected and used for the purpose, possibly based on “*trial and error*” experiments by the people over a period of time. Interviews with the salt makers did not throw light on this.

It is very clear that when the plant grows on the water banks, there is a direct uptake of minerals from water through soil. On contrary, if the plant grows away from the bank, then the minerals get absorbed from the soil, which in turn has mineral exchanges with water bodies. Needless to say, the flowing water certainly has leached minerals from various points of contact. More than seawater, spring as well as flowing waters are expected to contain more minerals because they could dissolve more mineral when they rise or flow and come in contact. Over and above, if the springs are hot or geothermal, then they contain much more dissolved minerals because at high temperatures the solubility of most of the inorganic salts is high. If this is the case, then it is really confusing to note that the people used only certain species for making ash and salt. In general, the people of PNG and Amazonian base, due to tough and rugged geographical terrains involved in the landscapes, probably could not move around and find such waters. It has been mentioned^[40] that the salt extraction through reduction of soils results in more sodium concentration, while that of the plants results in more potassium concentration. This was a scientific conclusion based on thorough data collection, experimental verification and data analysis. Such conclusions were not known to the traditional and tribal people living in interiors of the jungles. In that case how and why were they able to select plant species for fulfilling their salt diets is a puzzle. Godelier^[4,13,18,19], who lived with the Baruya people of PNG for some time and completed several scientific studies about them, has not mentioned any of the “*trial and error*” experiments undertaken by the people to choose the right salt source. Their methods include (a) collection and direct evaporation of the flowing water; and (b) drying, burning and working up of randomly collected plants. The reason for this doubt is that the organic parts of the plant are usually decomposed and volatilized when burnt, and only the non-volatile inorganic constituents remain in the salt. While people in Fiji and Central Borneo used either seawater or saline spring water, and it is very puzzling to note that the people in PNG who are in close proximity used only certain species of plants using laborious techniques and equipments. In search of answers, Enokakuio *et al.*^[41] have extracted the vegetal salts from randomly selected thirty different plant species that are commonly found in the Colombian Amazonian rainforest and the results obtained showed 0.9%-29.4% CO_3^{2-} , 1.1%-26.1% Cl^- , 7.9%-49.6% SO_4^{2-} , 0.01%-0.39% Na^+ and 28.2%-44.6% K^+ . These values are quite comparable with those obtained in the current study that were obtained from regular plants used for the salt production. This clearly suggests that any plant species, for that matter, could be used for the

salt production using traditional methods. This leads to a new question, why do various groups used a particular type of plant species? The three most probable reasons could be: (i) following the family tradition for generations, (ii) easily accessible, and (iii) their abundance.

Earth is the main source of minerals and other essential nutrients. In most of the cases, the Earth (soil) is present as an intermediate entity between plants and water bodies, and if water acts as a ‘messenger’ between plants and soils by actively dissolving the soluble minerals and taking them into plants (through absorption by the roots) and into water bodies like lakes, streams, *etc.* (through leaching process), then, it is more justified that water can be directly used as a convenient source for getting salts. A simple study on this would shed more light whether plants and plant ashes are required for making such salts. To support these arguments, salt composition of plant tissues were found to be dependent on their growth conditions and plants growing on brackish waters were used preferentially as a source of salt^[42,43]. The level of acidity or alkalinity of a soil greatly affects the level of availability of soil nutrients^[44] and hence the mineral content of a plant. It should be also noted that certain plants absorb certain ions more readily and easily than the others^[45] and this depends on the mobility or transportation of ions, solubility of ions, absorption by plants, availability in the system, mechanistic details and type of the plant, climatic conditions, *etc.* Depending upon the composition of saline water, ion toxicities may arise due to the predominant presence of a specific ion or nutritional deficiencies may arise due to either lack of certain cations and anions and/or competition effects among certain cations and anions^[46].

It is quite common that potassium ions are found in plants in high levels because this ion is considered as seventh most abundant element in the world and third major plant nutrient required for its proper growth and nourishment and thus the mechanism is such that almost all the plants naturally absorb it. Agricultural soils were estimated to contain around 2-30 tons of potassium per acre, and potassium is present in all the systems: humans, plants, animals, rocks and minerals, lakes and oceans, *etc.*^[47]. Almost all of the common potassium salts are highly soluble in water^[28], making the uptake easier. Phosphate (where phosphorus is the second major plant nutrient) has only been rarely reported^[12,17]. It should be noted that while alkali metal phosphates are soluble in water, alkaline earth phosphates are either poorly soluble or insoluble^[28].

4.13. Comparison with recommended daily intake (RDI)

It is further important to compare the values with the prescribed RDI to check if the salts are safe to consume or capable of producing any health issues^[48-50]. The reason for this is that the author has seen salt makers consuming a palm full of salt in one gulp (~10 g). So, assuming that the people on an average consume ~10 g of the salt each day (through cooking, consuming raw salt at times and consuming as a medicine with mashed ginger, cucumber and other herbs), the minerals content were calculated and compared with the RDI values. The values in the table for the seven salts were calculated using the following

formula, where 'x' is the percentage of weight of a particular element present in a salt.

$$\text{RDI (mg)} = \frac{x \text{ (g)}}{100 \text{ (g)}} \times 10 \text{ (g)} \times 1000 \text{ (mg/g)}$$

It is clear that most of the elements present in the salt seem to have no threat to the human consumption, except for the potassium content which exceeds the RDI in Aseki and Sepik river salts that coincidentally are deliquescent too. Iron content in Kerowagi salt was found to be marginally higher than the RDI value. Even if the amount of salt consumed per day gets doubled to ~20 g, all these elements are found to be well within the RDI limits but for the potassium and iron ions that certainly is a concern from the consumers' point of view. Large doses of potassium may cause stomach upsets, intestinal problems or heart disorders and iron doses > 20 mg may cause stomach upset and constipation or blackened stools[48-50].

While all the seven salts have low concentrations (< 0.001%-1.341%) of s-block metals (Li, Na, Mg, Ca, Sr and Ba), p-block semi-metal (B) and d-block metals (V, Mo, Mn, Fe, Cu and Zn), their potassium content is the most common and found to be present in abundance (21.64%-45.02%). The study brought out strong correlations between deliquescence and the nature of anions present, as well as solubility and conductivity. At higher percentage of ions ($\text{CO}_3^{2-} + \text{HCO}_3^- + \text{OH}^-$) greater deliquescence is noticed. In addition, with the solubility the electrical conductivity has increased substantially. It was realized that during the past, people actively involved in salt production and considered it as a big, routine and artistic activity. But, at present, these salts are being prepared remotely in certain interior villages, for their own consumption and donation to extended families, relatives and friends because readymade salts containing adequate quantities of iodide and fluoride are hygienically and industrially prepared and commercially available. Hence, in future, according to the authors' views, this traditional salt making and consuming practices would gradually diminish and finally perish due to vital factors like deforestation, population increase, urbanization of the interiors, educational priority, thriving for a high quality life, urge in quick earning, to name a few.

All the seven salts were found to be safe to consume, but Aseki and Sepik salts were found to contain high and marginally high potassium respectively (4502 and 4159 mg; 3568 mg) compared to the RDI value (3500 mg). Hence, daily consumption of such salts would lead to some biological problems as mentioned earlier; however they are fit for occasional consumption.

Conflict of interest statement

I declare that I have no conflict of interest.

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