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Occurrence and properties of natron in the Miocene lacustrine Beypazarı basin, Turkey

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With 10 figures and 3 tables

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Abstract: Natron, $\text{Na}_2\text{CO}_3 \cdot 10\text{H}_2\text{O}$, is a rarely forming evaporite mineral, which precipitates under the unique physicochemical conditions of Na wt. %, temperature and HCO_3/CO_3 . Natron is observed in the upper parts of the lower trona level of the Beypazarı Neogene volcanosedimentary sequences, where exist two trona beds between 260 m and 285 m depths. In this study, natron and some other soda minerals were examined by detailed geochemical, physicochemical and experimental studies. Euhedral natron crystals were determined sporadically within sedimentary beds. Based on our studies, it is postulated that white, soft and platy-fibrous natron minerals formed in the microenvironments within the rhythmic series of marls and claystones, are precipitated from pore solutions under the conditions of low temperature and high CO_3/HCO_3 ratio during the end of the first precipitation of the trona deposits. The contents of the trace elements were measured using wet chemical analyses; XRD, DTA and SEM techniques were performed, and the results were evaluated based on the formation conditions of the mineral.

Key words: Natron, REE, evaporate, soda minerals, Beypazarı, Turkey.

Introduction

Natron and other soda minerals are perhaps the most characteristic of the non-marine evaporite minerals, which precipitate under an unique basin condition. Their occurrence and mechanism of precipitation are also challenging from low temperature aqueous geochemistry point of view. Natron does not have any economic value, but it has scientific importance, like

trona. Generally, natron ($\text{Na}_2\text{CO}_3 \cdot 10\text{H}_2\text{O}$) has been observed together with thermonatrite in some localities of trona deposits (EUGSTER & SMITH 1965, SUNER 1989, 1991 a and 1993). Natron shows the transition to thermonatrite as a result of an increase in temperature and a decrease in water activity. Natron can form under the effect of atmospheric conditions in evaporative environments related to the chemistry of concentrated solutions, like playalakes, together with trona ($\text{NaHCO}_3 \cdot \text{Na}_2\text{CO}_3 \cdot 2\text{H}_2\text{O}$), nahcolite (NaHCO_3), gaylussite ($\text{Na}_2\text{CO}_3 \cdot \text{CaCO}_3 \cdot 5\text{H}_2\text{O}$), thermonatrite ($\text{Na}_2\text{CO}_3 \cdot \text{H}_2\text{O}$), halite (NaCl), burkeite ($2\text{Na}_2\text{SO}_4 \cdot \text{Na}_2\text{CO}_3$), wegscheiderite ($\text{Na}_2\text{CO}_3 \cdot 3\text{NaHCO}_3$) and thenardite (Na_2SO_4).

Determination of natron is significantly important, because it indicates the presence of industrial raw material deposits, such as halite and trona, which can not be easily determined by field geologists without drilling data. The CO_2 content of the gas phase equilibrated with the assemblages nahcolite + trona + solution, nahcolite + trona + halite + solution, and trona + thermonatrite + solution has been determined experimentally at the surface conditions (EUGSTER 1966). He concluded that the CO_2 contents of the paleoatmospheres must have been within the range specified by the natron + solution assemblages between three times and $\frac{1}{8}$ of the CO_2 content of the today's atmosphere. There are three major natural sodium carbonate (trona) deposits in the world. These are: Green River Formation in Eocene age from Wyoming (BRADLEY & EUGSTER 1969), Searles Lake of Pleistocene age in California in the U.S.A. (EUGSTER & SMITH 1965) and Lake Magadi of Pleistocene age to Recent in East Africa (BRADLEY & EUGSTER 1969).

Natron minerals are formed within volcanosedimentary sequences in Turkey (SUNER 1989). The soda minerals, determined in the Neogene volcanosedimentary sequences, may be classified in two groups. One of these is the World's second largest trona deposits with 200 million ton reserve and the other group is consisted as rare forming soda minerals, which are accumulated within the upper part of the same stratigraphic sequences with very small occurrences. In this paper, mineralogical and geochemical characteristics were investigated; the physicochemical parameters under which natron has been formed and deposited were discussed. The possible relations to other observed evaporative species in term of formation conditions were researched. The purpose of this paper is to discuss physicochemical conditions of natural natron occurrence in the Beypazarı basin and to document its mineralogical properties.

Regional geology

Beypazarı – Çayırhan lacustrine basin has an economic importance with the respect to the presence of large trona deposits (Fig. 1). Natron and other rare-forming evaporative minerals, such as thermonatrite, nahcolite, pirssonite, gaylussite, shortite, had been occurred within the same stratigraphic sequences which are interbedded with mainly tuffs, tuffites, marlstones, claystones and bituminous shale (HELVACI et al. 1989, SUNER 1994 a and 1994 b).

The rocks observed in this lacustrine basin are in the following groups (Fig. 1): a) Paleozoic metamorphic basement rocks, b) granodioritic rocks, c) Jurassic – Cretaceous carbonate rocks, d) Cretaceous flysch and limestones, e) Upper Cretaceous ophiolitic rocks, f) Eocene – Paleocene aged clastic sedimentary rocks, g) Neogene volcanics, h) Miocene lacustrine rocks, i) Neogene clastic sedimentary rocks (HELVACI et al. 1989, SUNER 1993).

Natron was found in the Miocene lacustrine rocks that were consisted of six units ranging in thickness from 60 m to 350 m (Fig. 1). All these units had been deposited conformably, and natron was determined on the upper parts of the lower trona horizon within the same sequence. During the time of Miocene deposition, extension and compression regimes in different scales were dominant. Obviously, the progressive tectonism had effected the formerly deposited accumulations and consequently, they had been dissolved and reprecipitated in the form of different chemical compositions within, especially, the rhythmic claystones / marlstones units where cavities and cracks were commonly observed. Therefore, these evaporative accumulations were deposited in the form of many thin beds or lenses. Under the effects of burial conditions of the lacustrine sediments, the soda minerals lost their water contents from 10 to 1 molecules in crystal structures during the dehydration process, controlled their paragenesis, resulting in the thermonatrite formation. During this progressive process, many thin volcanosedimentary sequences had been formed interbedding with trona levels. Natron and other rare-forming soda minerals had been also formed in different parts of these sequences, depending on their chemical compositions of the concentrated solutions.

Material and methods

The first determination of natrons on core samples obtained from 260–285 m depth, containing trona and claystones – marlstone complexes. Core

samples were taken from 300–400 m depth in the Beypazari basin. No brine (borehole) temperature is available for the levels of core samples that were collected. First, samples were studied under binocular microscope and were tested by using XRD, DTA and SEM techniques. The chemical analyses were also performed by wet chemical, gravimetric, colorimetric and spectrometric methods.

In this study, Philips 1140 model XRD was used and the XRD investigations were performed under the conditions of $2\theta = 1^\circ$, Ni-filter and $\text{CuK}\alpha$ radiation. For the DTA studies, the Pt–Pt/Rh thermoelement, normal atmosphere, the $10^\circ\text{C}/\text{min}$ heating speed and 2.5 mm/min recorder speed were used. SEM investigations were also conducted and Jeol type JSM–35 Electron Microprobe was used. Na_2CO_3 analyses were conducted by using E–536 and EA–21 combined electrodes, 0.1 N HCL and Na_2CO_3 as the reference solution. During the analyses, the contents of Cl, I, F, Br, Cs, Fe, K, Ti, Mg, Ca, Rb, Ba, Sr, Li, Si, Al, B, As and SO_4 were measured.

Mineralogy and petrography

Natron was found together with trona, as the main mineral and occurred in the upper parts of trona crystals in the basin. The other rare forming occur-

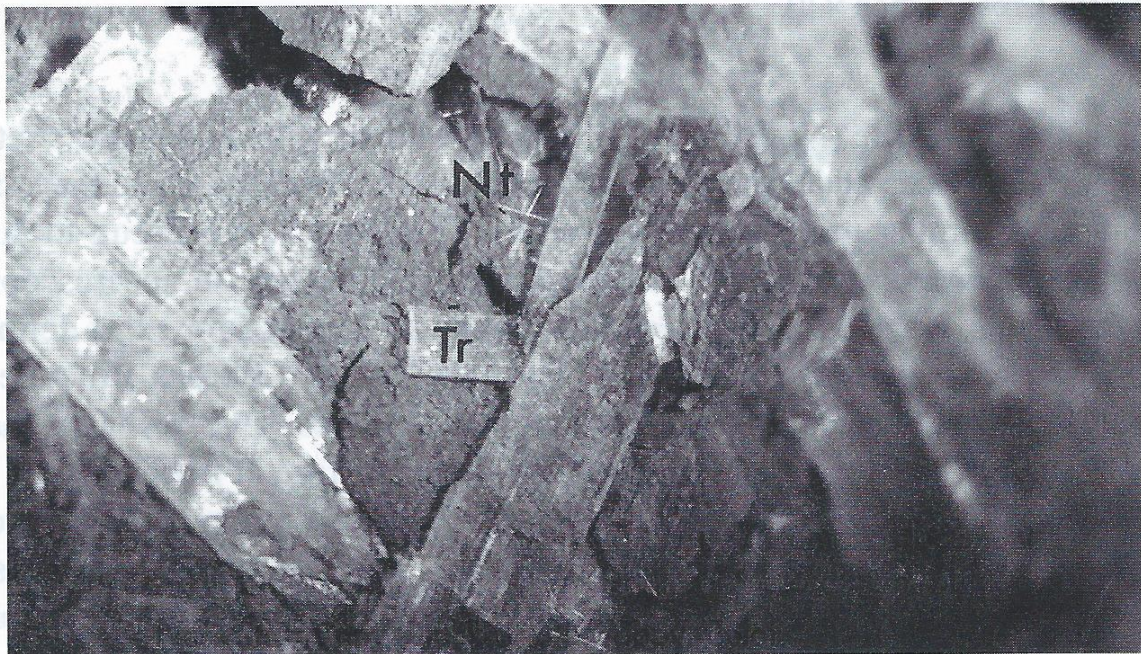


Fig. 2. Natron crust on the pure trona hand-specimen under the polarizing microscope.

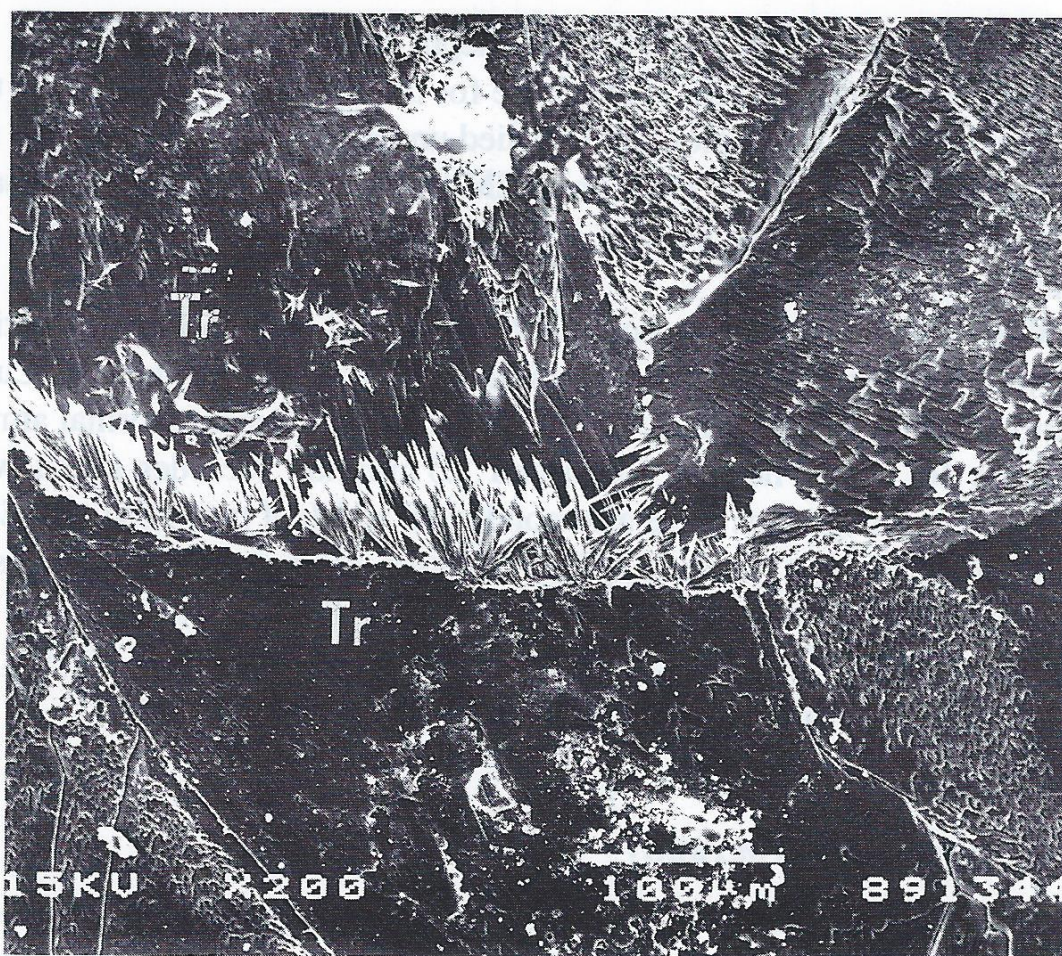


Fig. 3. Fringe of acicular natron (Nt) crystals exhibit on radially fibrous trona (Tr) crystals. Cross section $\times 10$.

rence, thermonatrite ($\text{Na}_2\text{CO}_3 \cdot \text{H}_2\text{O}$), was also found within the same stratigraphic sequences under the similar depositional environments and they exhibit very similar morphologic properties. Therefore, it is difficult to distinguish between natron and thermonatrite, particularly in macroscopic studies (Figs. 2 and 3). Hence, detailed studies are required for exact definition. These minerals are white and they have soft and fibrous structures. The studies performed under polarizing microscopes have revealed that they had been formed as a thin platy cover on the surface of trona uystals.

The results of XRD and DTA studies are presented in Figs. 4 and 5, respectively. DTA analyses are more definitive than XRD studies, because their data about dehydration contents are determinative in the natron – thermonatrite distinctions. The XRD and DTA data are presented in Table 1 and 2, respectively. Two XRD peaks observed at 29.35 and 30.46 ($2\theta^\circ$) are the most distinctive points and DTA remarks of natron are characterized by a

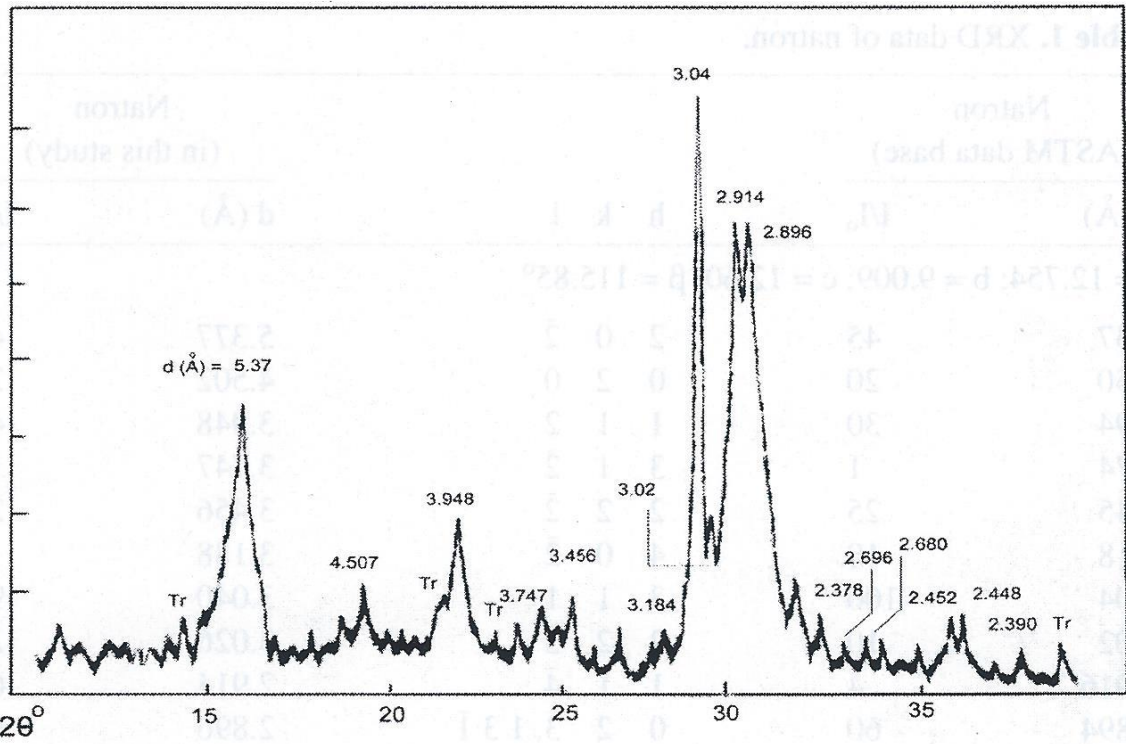


Fig.4. XRD pattern of natron mineral.

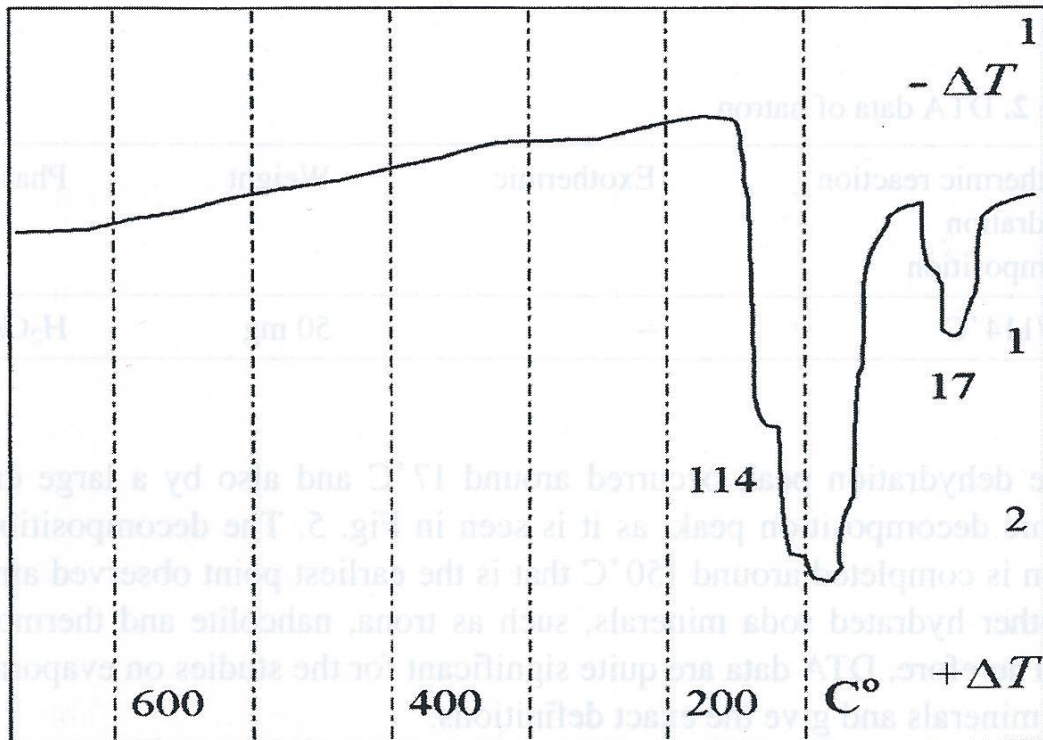


Fig.5. DTA curve of natron mineral.

Table 1. XRD data of natron.

Natron (ASTM data base)						Natron (in this study)	
d (Å)	I/I ₀	h k l				d (Å)	I/I ₀
a = 12.754; b = 9.009; c = 12.60; β = 115.85°							
5.37	45	2 0 $\bar{2}$				5.377	45
4.50	20	0 2 0				4.502	22
3.94	30	1 1 2				3.948	40
3.74	1	3 1 $\bar{2}$				3.747	10
3.45	25	2 2 $\bar{2}$				3.456	20
3.18	10	4 0 $\bar{2}$				3.188	8
3.04	100	3 1 1				3.040	90
3.02	10	2 2 $\bar{3}$				3.026	20
2.916	4	1 1 $\bar{4}$				2.914	60
2.894	60	0 2 3, 1 3 $\bar{1}$				2.896	68
2.742	2	1 3 1				2.738	12
2.697	12	2 2 2				2.696	7
2.684	12	4 0 $\bar{4}$				2.680	8
2.448	2	3 3 $\bar{1}$				2.448	20
2.399	6	0 2 4				2.390	7

Table 2. DTA data of natron.

Endothermic reaction	Exothermic	Weight	Phase
Dehydration Decomposition			
17 °C/114 °C	–	50 mg	H ₂ O/CO ₂

single dehydration peak occurred around 17 °C and also by a large endothermic decomposition peak, as it is seen in Fig. 5. The decomposition of natron is completed around 150 °C that is the earliest point observed among the other hydrated soda minerals, such as trona, nahcolite and thermonatrite. Therefore, DTA data are quite significant for the studies on evaporative soda minerals and give the exact definitions.

As the result of SEM investigations, characteristic fibrous and acicular natron crystals were examined. As it is noticed in Fig. 6, equal dimensions

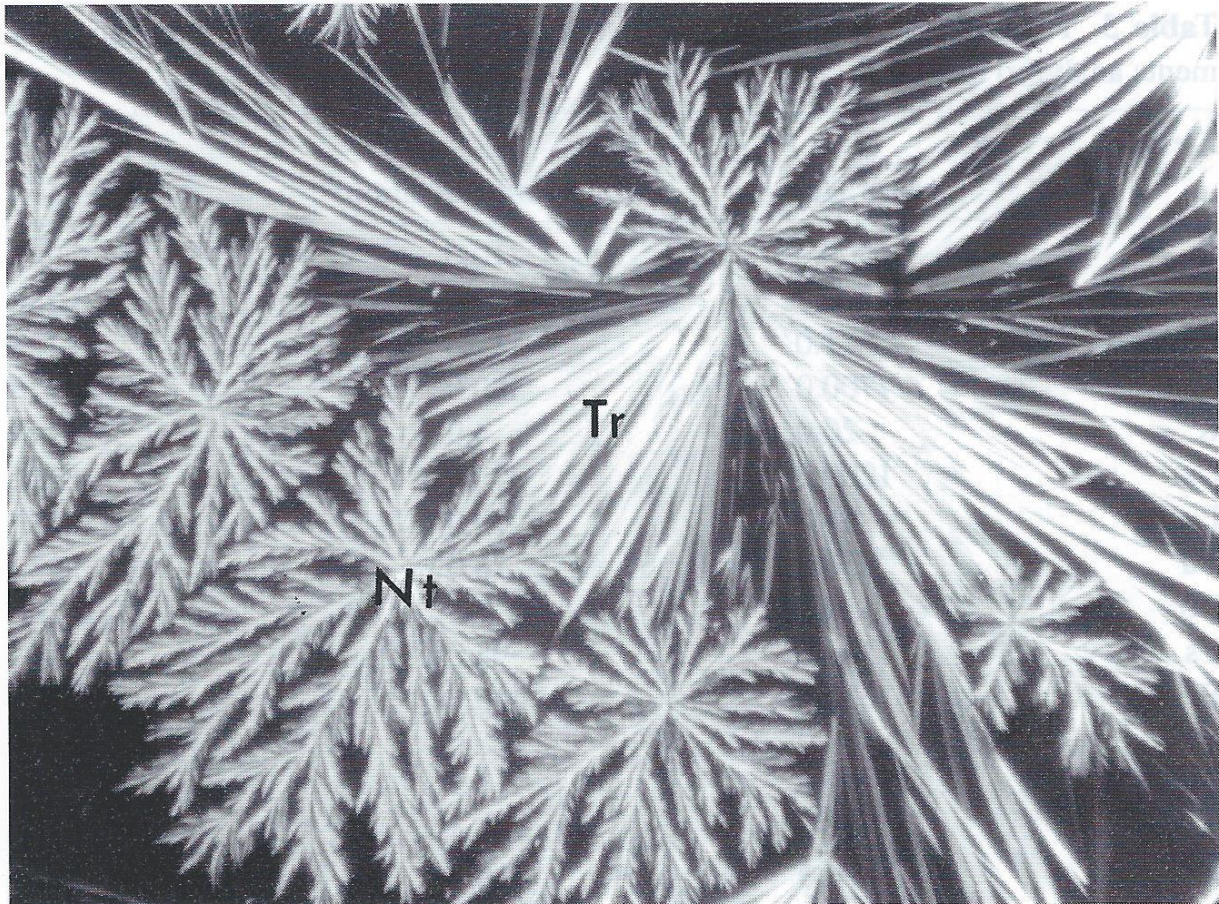


Fig. 6. SEM images of natron (Nt). Fibrous and acicular crystals on trona (Tr) planes.

are commonly indicating the presence of rapid forming conditions. This point is important in terms of the paragenetic evolution for the basins. The minerals were observed generally are in the cracks and on the thin microstructures of trona crystals (Fig. 6).

Geochemistry

Geochemical studies focused particularly on the trace element contents that are more definitive in the genetic interpretations, especially for the studies, which deal with the formation conditions and genetic discussion of the rare forming evaporative soda minerals. Evaporate minerals that may easily change in their composition, are subjected to the alteration and weathering processes. A series of evaporative minerals can be found together with the same volcanosedimentary lithologies. For this reason, many evaporite occurrences were discovered in this basin and trona + nahcolite + thermonat-

Table 3. The average values and standard deviations of the major and trace elements analyzed in natron.

Element	X (Average)	S (Standard Deviation)	S _k (Skewness)
Si	1500.0	210.0	-0.285
Al	320.0	52.0	0.288
Fe	71.0	12.0	0.125
Mg	7700.0	380.0	0.276
K	20.0	4.9	0.244
Ca	1200.0	210.0	0.21
P	85.0	19.0	-0.17
Ba	550.0	78.0	-0.23
Rb	10.0	2.9	-0.20
Cs	700.0	95.0	-0.16
Br	1650.0	410.0	-0.11
Cl	580.0	112.0	-0.16
B	23.0	5.7	-0.26
As	310.0	6.4	-0.17
F	5600.0	710.0	0.08
I	1200.0	215.0	-0.14
Li	5.0	1.4	-0.23
Sr	220.0	4.8	-0.18
Ti	3.0	1.2	-0.3
SO ₄	10500.0	230.0	0.26

rite + natron + pirssonite + gaylussite paragenesis were determined by the detailed mineralogical investigations.

During the laboratory experiments, major and trace elementary analyses were done in order to obtain a large data set for genetic interpretations. The statistical parameters, the average values, standard deviation and skewness coefficients were given in Table 3. Because of the paragenetic assemblages of trona and natron, which are observed in macroscopic scale, the elementary composition of these minerals were compared with the aim to investigate possible similarities in genesis and formation conditions, and the results are presented in Fig. 7. The results of these studies reveal that there is a strong similarity in term of elementary composition and the trend, which shows evidently the relationship between these two minerals. Furthermore, from the statistical parameter point of view, the similar trace element con-

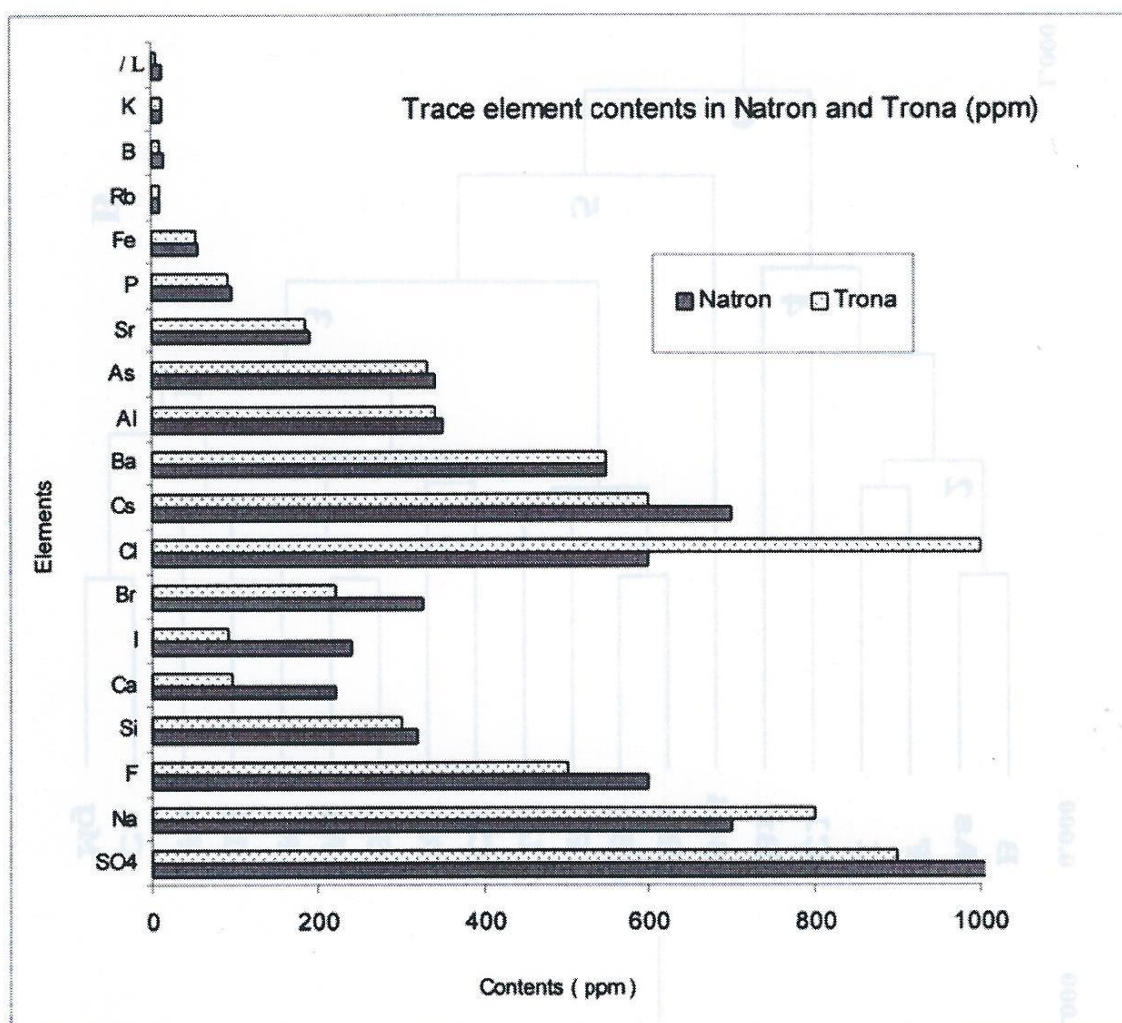


Fig. 7. Diagrams showing the distributions of trace element contents determined in trona and natron samples.

tents are noticed. This observation is also supported by the results of cluster analysis, as it is seen in Figs. 8 A and 8 B.

Physicochemical parameters

Temperature, and activities of CO_2 and of H_2O are the critical parameters, which directly control mineral phases and their amounts (EUGSTER & SMITH 1965, HARDIE 1968, BRADLEY & EUGSTER 1969, HARDIE & EUGSTER 1970, EUGSTER 1970, 1979, EUGSTER & MAGLIONE 1979, EUGSTER et al. 1980). The values of $a_{\text{H}_2\text{O}}$ and a_{CO_2} in the brine, where soda minerals are in equilibrium, are controlled by the composition of the brine. With increasing salinity, the magnitude of $a_{\text{H}_2\text{O}}$ decreases in general. The magnitude of a_{CO_2} does not directly correlate with salinity, but is controlled by the amounts of

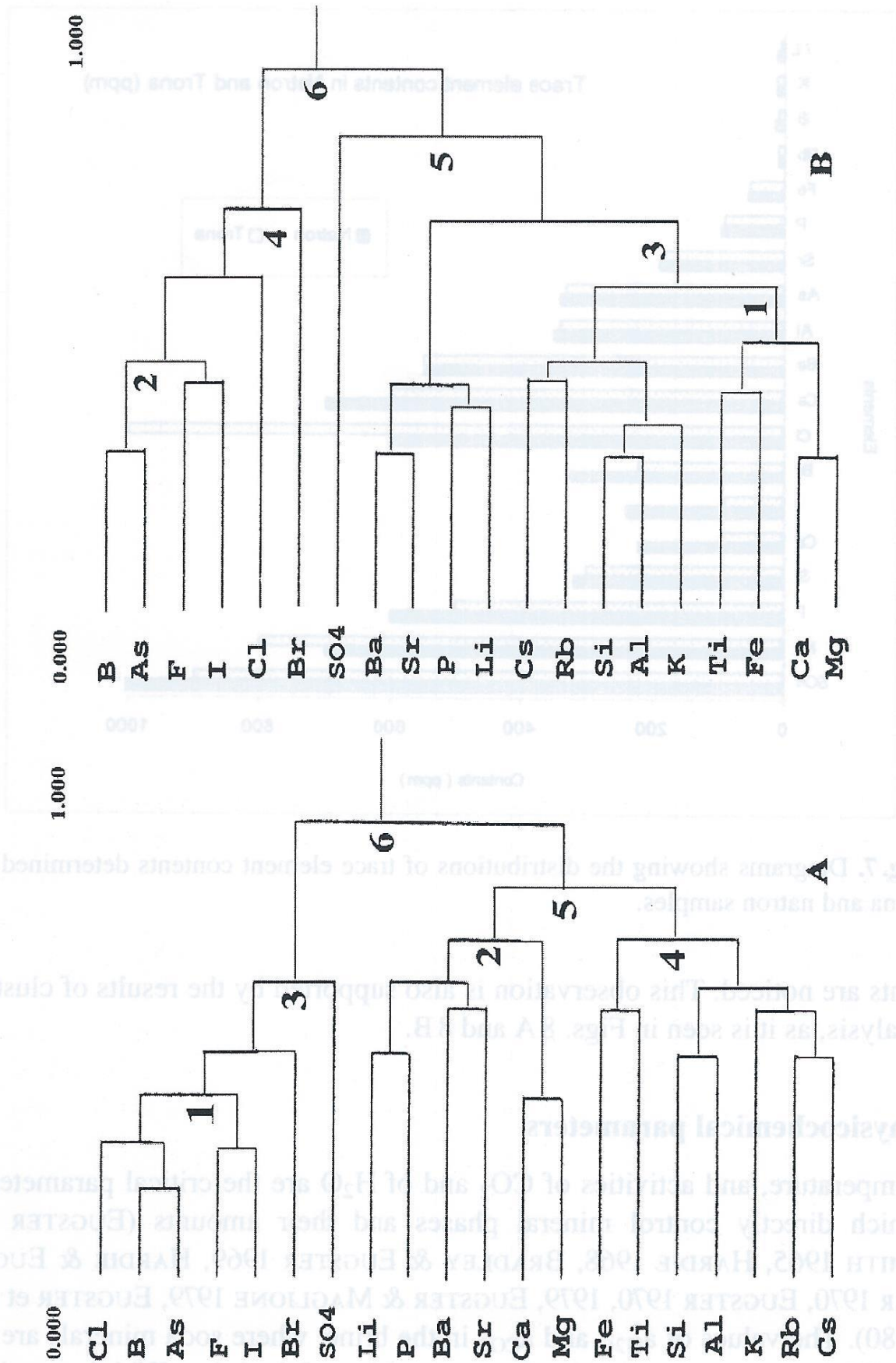


Fig. 8. The cluster results performed on the samples of natron (A) and trona (B).

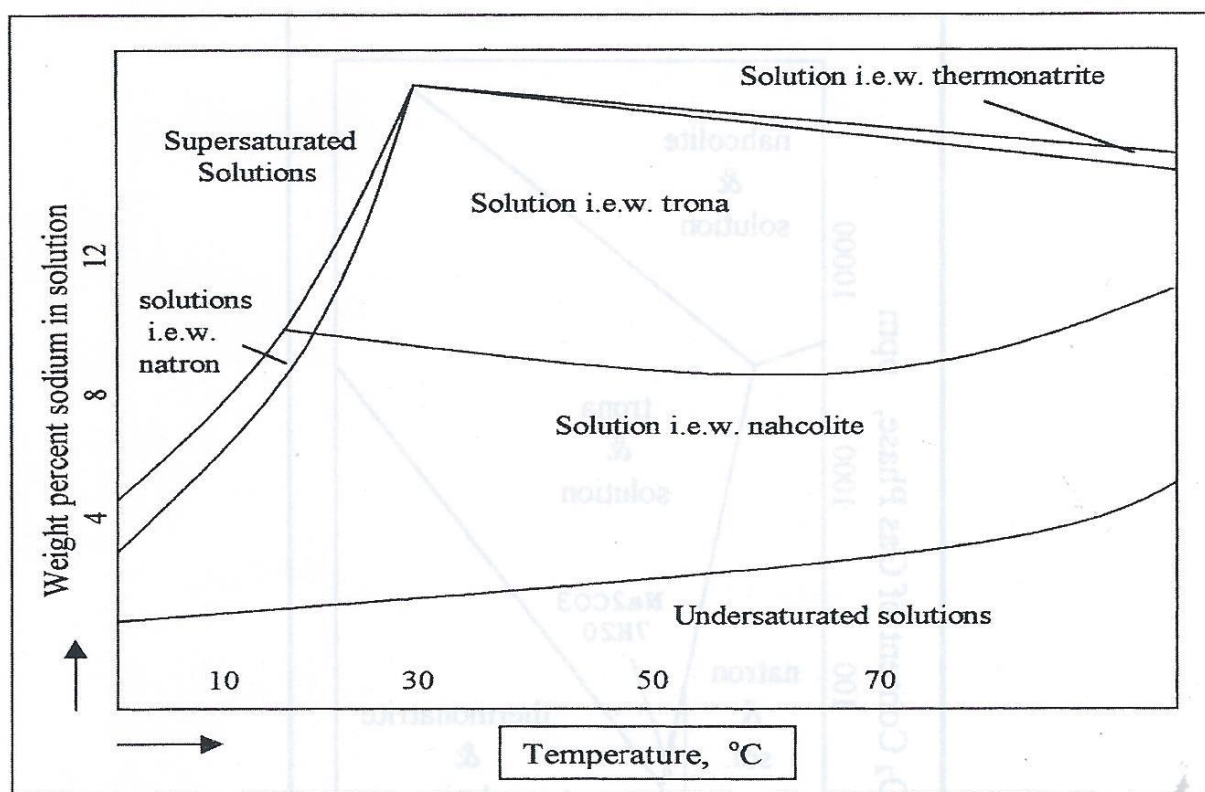


Fig. 9. Natron–Trona–Nahcolite–Thermonatrite stability areas as function of Na wt. % and temperature at 1 atm pressure (BRADLEY & EUGSTER 1969).

HCO_3 and CO_3 present in the brine. Changing of these parameters are resulted in a series of transformations and consequently, the occurrence of new mineral paragenesis were given by various investigators (EUGSTER 1966, BRADLEY & EUGSTER 1969), as it can be seen in Figs. 9 and 10, where the stability areas of natron and trona together with nahcolite and thermonatrite are remarked.

The stability area of natron is controlled mostly by a change in temperature (EUGSTER 1970, EUGSTER & MAGLIONE 1979). An increase in this parameter is concluded that the natron–trona or natron–thermonatrite transformations are depending on activity of CO_2 . Fig. 9 points out that this change is generally in the favor of trona more than thermonatrite, due to the differences in the boundary properties of the two minerals (i.e., the boundary of natron–trona is slightly sharper than that of the latter). For this reason, natron–trona assemblage is commonly observed and they have been determined together, as we have found in the Beypazari basin.

EUGSTER (1966) experimentally determined $\text{H}_2\text{O}-\text{Na}_2\text{CO}_3-\text{NaHCO}_3$ relation and he demonstrated the role of temperature and P_{CO_2} in natron–trona–thermonatrite–nahcolite precipitation at 1 atm pressure and 25 °C tem-

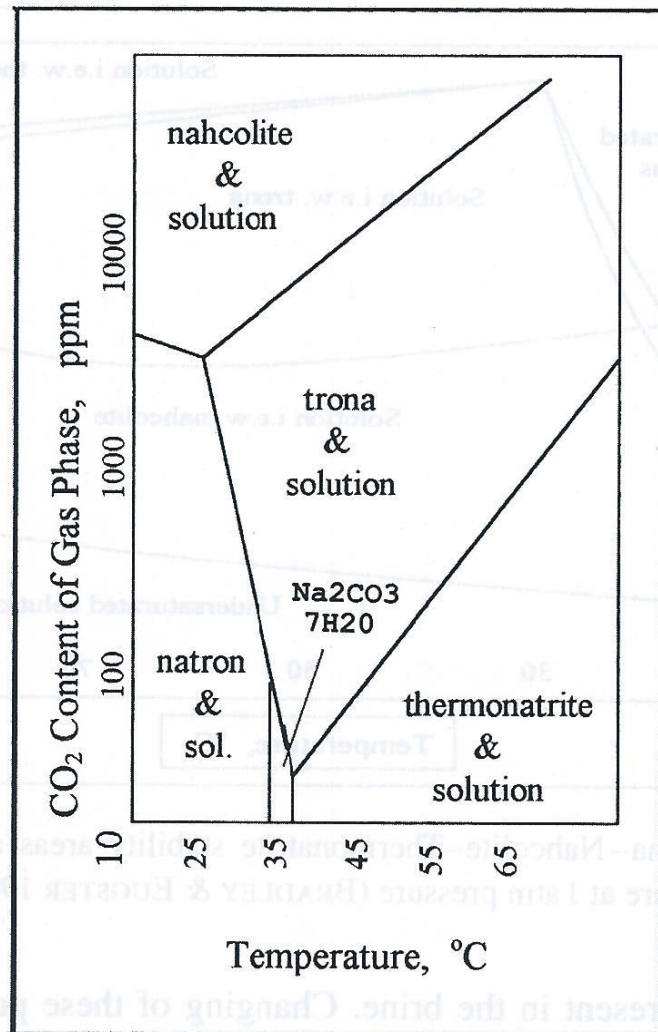


Fig. 10. Natron–Trona–Nahcolite–Thermonatrite stability areas as functions of CO₂ (ppm) and temperature at 1 atm pressure (BRADLEY & EUGSTER 1969).

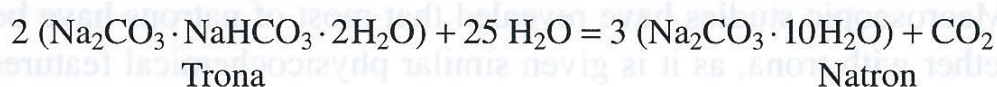
perature. Fig. 10 shows that trona–natron–Na₂CO₃·7H₂O–solution boundary lie about 1/8 of the CO₂ content of today's air at 32–35 °C temperature ranges. Trona, nahcolite and thermonatrite have been reported in literature (REITSIMA 1980 and SUNER 1991 b). However, very little data are available for natron occurrence and experimentally made natron is less studied among the soda minerals. In addition, Na₂CO₃·7H₂O mineral, like other researchers, is not found in this study. EUGSTER (1966) reported that natron is supposed to crystallize from carbonate brines in the winter, and probably present in some salt lakes of Hungary, Egypt and perhaps in Owens Lake, California and Alkali Valley, Oregon.

Of course, the most important argument on the precipitation of soda minerals is that whether equilibrium was existing with respect to CO₂ between the atmosphere and the Miocene brine or not. If we assume that the argu-

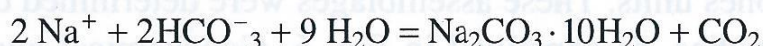
ment is applicable to our case during the time of trona deposition, then we can use Fig. 10 to place limits on the CO₂ content of that Miocene atmosphere. Assuming that a reasonable average temperature for evaporation was 25–35 °C; trona and natron will precipitate when the CO₂ content of the Miocene atmosphere is not more than 3 times and not less than one-eighth that of the CO₂ contents of today's atmosphere.

Trona is the main mineral in Figs. 9 and 10, indicating its property of having the highest stability area among the other evaporative carbonate species observed in the studied basin. Trona can easily form under the effect of atmospheric conditions and also of the partial pressure of CO₂ (REITSIMA 1980), which provides the properties of forming in actual lacustrine lakes, as it is precipitated today in the lakes Chad and Magadi in Kenya (EUGSTER 1970, EUGSTER et al. 1980). On the contrary, natron formation is restricted in term of Na wt. % in Fig. 9, although it can also form rarely under atmospheric conditions.

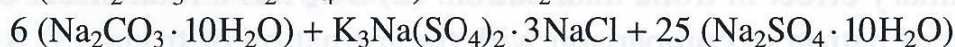
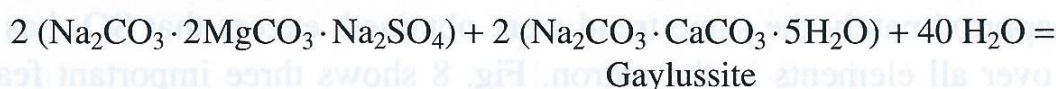
Natron may form primarily and also secondarily from the solutions or from evaporative minerals such as early formed carbonates and bicarbonates, which have been precipitated according to the composition of the solutions during the opening periods of the Beypazarı basin (SUNER 1989, 1991 a, 1991 b).



or



Not only Na-bearing carbonates but also Ca/Na double carbonates/bicarbonates/sulphates may be responsible for natron formation, as it is explained as follows:



In general, all Na-bearing evaporative minerals, especially trona, natron, thermonatrite are very soluble complexes. They may easily decomposed under the effect of meteoric conditions and then, they turn into liquid phases.

Consequently, it is difficult to discover them by field research, without drilling core sample. Therefore, in this case, they had been protected within the sedimentary sequences, such as claystones, marlstones, tuffs and tuffites, and many rare forming minerals may form very scatteredly in these units.

Conclusions

All natron occurrences are observed in the form of thin crust either on the upper part of the lower trona levels or within claystones–marlstones units. Natrons are observed very rarely in the whole trona deposits and they are also determined as a result of the studies performed on the core samples. The mineral is soft and white in color, and it carries fibrous and acicular structures. Because of large amount of water molecule, i.e. $10\text{H}_2\text{O}$, it can form under the effect of high activity of H_2O . Due to the presence of many trona levels and also due to the actual tectonism, necessary Na and CO_3 ions are present in the concentrated solution. On the other hand, $\text{CO}_3 / \text{HCO}_3$ ratio is equal to one in the trona structure, which is another critical point in term of the formation conditions. Atmospheric conditions are not a limiting factor during the crystallizations of natron, as discussed above.

Macroscopic studies have revealed that most of natrons have been found together with trona, as it is given similar physicochemical features. Trona–natron paragenesis is postulated and some of natrons were observed within claystones-marlstones units. These assemblages were determined during the studies of SEM studies, as explained in Fig. 6. Geochemical analyses focused on the contents of the major and trace elements exhibiting a similarity between these two minerals, as it is presented in Fig. 7. The cluster analyses were performed and are presented in Fig. 8. A high positive correlation and approximately the same trend were obtained, except that SO_4 has clustered over all elements in the natron. Fig. 8 shows three important features; (1) volatile elements are more representative in Fig. 8 A than 8 B for the influences of calc–alkaline volcanic activities and thermal solutions, indicating the primary effect in trona than natron. (2) SO_4 has a final effect on the formation of natron in comparison with the formation of trona. (3) At the time of closure stage of the basin, oxidizing atmospheric conditions became more dominant during the occurrence of natron formation, because significant volume of fresh water had entered into the basin, covering up the upper level of trona deposits.

In the Beypazarı basin, natron was determined on the upper part of the lower trona horizon. On the other hand, the upper parts of the upper trona horizon are covered mainly by nahcolite (NaHCO_3), which is one of the important minerals in carbonate-rich lakes and natron was not observed within these levels. This observation is interpreted as a result of the different evaporative conditions, the existence of time interval between the two main formations and also decreasing in pH values upward sequence.

At the end of the first evaporation period, natron has been deposited secondarily from the solutions generated from tronas because of the increasing ratio of $\text{CO}_3 / \text{HCO}_3$, activity of H_2O and temperature. In addition, natron has found the occasion of forming within a very limited period because of the incongruent melting property of trona. During this stage, the concentrated solutions have contributed to natron crystallizations in the form of pore crystals under the effect of diagenesis.

At the closing time of the basin, natron could not formed because of the increase in the HCO_3 content of the solutions. Therefore, upper parts of the Beypazarı basin were covered by nahcolite crusts, which are formed in low temperatures and under the effect of high CO_3 activity. No halite mineral is found in core samples. We assume that the absence of halite formation at 260–290 m depth is due to insufficient amount of Cl ions in the system. As the result of these studies, it is concluded that natron has been formed at the end of the first evaporation episode, due to the increasing ratio of $a_{\text{H}_2\text{O}} / a_{\text{CO}_2}$ and increases in $\text{CO}_3 / \text{HCO}_3$ ratio within the same volcano sedimentary sequences.

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