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RESEARCH ON NaCl SALINE AEROSOLS I. NATURAL AND ARTIFICIAL SOURCES AND THEIR IMPLICATIONS

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Abstract

This paper is the first of a series of new researches on the NaCl aerosols, which includes the bibliographical analysis and synthesis regarding the structure, the form and the sizes of the particles of the NaCl aeroions in correlation with the production procedures. There are also analyzed some practical applications pointed out both by some ancient Greek and Roman texts and by the present-day literature; at the same time there is underlined the existence, even from Antiquity, of an *ethnoscience* which led to the current knowledge in the *halotherapy* field. Concerning the artificial sources of the aerosols employed in the multiple-usage halochambers, there have been taken into consideration the most well-known procedures which are based on the *mechanical separation* or *erosion*, followed by the *physical dispersion* in the atmosphere; on the *breaking of the gas bubbles* in the sparging with air or other inert gases by means of the supersaturated saline solutions; on the *atomization of the saturated saline solutions* in the vacuum centrifugal air separators and the *carrying-away of the superficial particles* resulted from solvolyses and the consecutive anhydrizations of the structures from the surface of the halite blocks. These procedures will be included in the subsequent research.

Key words: aerosols, ethnoscience, halotherapy, NaCl, particle size, sources

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1. Introduction

The NaCl aerosols resulted from naturals sources (marine aerosols and saline aerosols from salt mines) or artificial sources (halochambers, saline devices or inhalers) are polydispersed systems with special nano-structural properties and with multiple practical implications (for example, in the prophylaxis and therapy of some respiratory affections, in the improvement of the parameters of the cardio-respiratory apparatus as well as of the psycho-neuromotory parameters, in the purification and the improvement of the quality of the atmospheric air etc.).

Depending on the *type of the source*, the *activity of the particles* and their *life cycle* as well as on the *environment conditions*, respectively, the atmosphere aerosols present a *dimensional distribution* and a somewhat steady *concentration* because of the difference between the *production speed* and the *loss speed*, all these being considered as a result of the condensation, coagulation, peptization, electroneutralization and sedimentation (destabilization) processes, respectively etc. (Sandu et al., 2003; Ştefan, 1998).

A special feature of these particles is their chemical composition, which varies in time and results from the interaction with the water dipoles and with other particles or ions, an interaction which is correlated with the form and size of the aerosol (Sandu et al., 2003; 2009a; 2010a). Thus, for therapeutic environments there are used gaseous

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sub-micronic micro-dispersions both as ”wet-aerosols” and ”dry aerosols” while for ambiental conditions there are employed especially the last ones generate the ”fresh air effect”.

Depending on the characteristics of the source and on the environment conditions in which they are obtained, respectively, the aerosols have variable forms, granulometries and concentrations. One source, especially an artificial one, is characterized by *three specific zones*, namely, the *active layer*, the *diffused layer* and the *residual or passive layer*, zones which are differentiated by the *activity* of the aerosols as well as by the concentration and sizes of the particles, respectively (Sandu et al., 2004a; 2004b).

The saline aerosols resulting from natural or artificial sources, used especially both for various treatments and for prophylactic purposes as well as for increasing the performances of the human subjects, must have a certain *activity*, i.e., a certain level of concentration and a certain well-controlled domain of granulometry, respectively.

Concerning the usage of the aerosols there is an interesting history that proves the existence of an ethnoscience, which has generated during some time periods various applications of the halite, salt marsh and saline aerosols. Moreover, this ethnoscience has led to a new branch, i.e. the halotherapy, very much used now-a-days, and having great development prospects in the future.

This paper presents a series of aspects connected with the structure of the aerosols, with the obtaining methods and with the practical implications known from Antiquity till the present times, data which are very important for the development of some researches regarding the halotherapy, researches which will be discussed in some future papers.

2. The size and form of the NaCl aerosols particles

From a chemical view point of view, the sodium chloride is a strong saline electrolyte with a high wettability and a limited solubility in water (35.7 g/100 g H_2O at 0°C and, respectively, 39.12 g/100 g H_2O at 100°C) or in other polar solvents; sodium chloride can exist, both in the liquid and gaseous environments, as crystalline nano-dispersions interacting with the dipoles of the dispersion medium which gives them a variable chemical structure, under a glomerural form, with a specifically negative charge (Sandu et al., 2004; 2009a; 2010a).

Since the particles have a negative charge, their *life cycle* varies from a few minutes to a few days, depending not only on a series of external or environmental *exogen factors*, i.e. humidity, temperature, pressure, light, presence of some dispersions with positive charge as well as of the organic ones, but also on some *endogenous factors*, i. e. the form, the side and the nature of the grains (powders), the method of obtaining or the type of the source, the production speed (capacity or the output of the source), the rate of coagulation or sedimentation etc. (Sandu et al., 2003; Ştefan, 1998).

Therefore, a specific characteristic of these nano-dispersions is the continuous structural modification due to the electrostatic and stereo-specific interactions with the dipoles and the aircations existing in the dispersion medium.

The dynamics of these processes is determined by the intensity of the exogen factors. In aqueous liquid mediums the size of the particles varies from the average diameter of the ”aquated ion” ($Na^+_{(aq)}$ and $Cl^-_{(aq)}$, respectively) up to some tens of microns of a cluster-type ionic nanostructure $[(NaCl)_n{\cdot}xH_2O]^-_{(aq)}$, the so-called ”solion” while in the gaseous mediums where there are only ”solions” with various diameters and structural forms, the size of the particles ranges from tenths of microns to tens of microns (Sandu et al., 2010). The various structure of the ”solion” particles in the gaseous mediums is due to the different degree of hydration with water dipoles and to their way of suprastructuring, taking multiple forms with spatial structures similar to snow flakes only at a nanostructural level. The multiple structural arrangement (Sandu et al., 2002) is considered to be the result of the compatibility between the cubic crystalline lattices of the sodium chloride and of the pentahydrol water $(H_2O)_5$.

Depending on their activity and life period, respectively, the aerosol particles have a somewhat steady dimensional distribution which is the result between the production speed and the loss by different processes. From this view point, they can be divided in five groups or dimensional levels which correspond to some various measuring methods and techniques; here they are.

- *small simple ions* with diameters under 0.5 nm which can be determined only in solution or smelt by electrochemical methods (Gulea, 1994);
- *Aitken particles* with diameters between 0.5 and 50 nm, detected with the *Aitken Particle Counter* (Junge 1963) or by means of the *mass spectroscopy* (Mitchell and Nagel, 2004);
- *average-size particles*, with diameters between 50 and 100 nm, detected with the *Laser (phase) Doppler System* (Mitchell and Nagel, 2004), *Angular Intensity Light Scattering* (Mulholland et al., 1985) and *Quasi-elastic Light Scattering* (Mitchell and Nagel, 2004) methods;
- *large particles*, with diameters between 100 and 1,000 nm, detected with the *Laser Diffractometer* (LD) (Mitchell and Nagel, 2004), *Electronic Microscopy Transmission* (MET) (Lettieri and Hembre, 1989) and *Electrical Sensing Zone* (ESZ) (Mitchell and Nagel, 2004) methods;
- *huge particles*, with diameters more than 1,000 nm (up to some tens of microns), detected with the *Electrical Mobility Analyser* (Kinney et al., 1991) and *Optical Microscopy/image Analyser* (Hartman et al., 1991; Hartman and Doiron,

1992; Lettieri et al., 1991; Thom et al., 1985) methods;

The classification of the NaCl aerosols according to the dimensional groups and to the form is shown in Table 1.

It is known a method and a device used for determining the concentration of NaCl aerosol in various natural and artificial halochambers (Pascu et al., 2009b). These were used in the study of different wet and dry aerosol sources (Sandu et al., 2003; 2004b), but also in the study of the influence of exogenous factors (Sandu et al., 2003; 2004a).

3. From ancient application to modern halotherapy

A series of comparative studies (Sandu et al., 2009a; 2010a), presenting the data obtained from ancient writings and from ethnographic surveys, have concluded that salt is being used with multiple applications (as natural brine or solid salt). Among these we mention: food conservation, treatment of different diseases, treatment of wood and animal skins and water purifying, applications which are used as traditional procedures in the countryside until today.

The other important objective of this study is to highlight the validity of the corresponding ancient and current practices, analyzed from the scientific perspective of biochemistry and biophysics. The continuation of the ancient halotherapeutic practices in rural Moldavia proves that the area under discussion still has a true ethnoscience in the field. Simple people in the Moldavian villages know the beneficial effects of salt in the treatments against certain diseases, without being interested in providing scientific explanations for those effects.

The use of the halite and brine in various therapies, as such or as powders and solutions (natural or artificial salt water) in ancient times and in the current traditional areas or in modern preparations (pomades, tooth pastes, soap, sticks, etc.) has a scientific explanation in the properties of the nanostructures. These nanostructures may be dissolved or dispersed in the usage systems whose negative charge is usually represented by what we may call ”solvions.” The latter become effective in the osmotic processes that influence the mechanisms of the dysfunction recovery. These actions begin at the epithelial surfaces but can also involve more specific metabolic processes.

In an organism salt performs a series of actions in accordance with the concentration level in the biotic system, with its mineral composition, with its retention and elimination capacity and with the coagulating capacity of the gelling systems. High concentrations of salt also have antibacterial, antimicrobial, and even antimycotic activities. This is how we explain, besides the therapeutic action, its various uses in food preservation (Chervinskaya, 2007; Hedman et al., 2006; Sandu et al., 2002; 2003; 2006; 2009a; 2010a). More details concerning these actions are given below:

- action on the nervous system through the ionic effects upon axons and neuronal cell bodies, reducing the pain (dental diseases, neuralgias, etc.);
- action upon mucosal surfaces, in particular their microbiotic systems, with effects extending to internal areas (such as salivary glands, middle and inner ear, sinus cavity, gingival zone, oral cavity, etc.) and physical lesions (including stings, bites, cuts, etc.).
- antimicrobial actions in cases of tonsillitis, sinusitis and otitis, as well as infections after bites, stings, etc. The mechanisms include the astringent effect (i.e., protein denaturation), inhibition of the microbial metabolism as well as the increased efficiency of the leukocyte system;
- antagonism of rheumatic processes, by its effects on the erythrocyte sedimentation rate (ESR) and by the modulation of the processes in the circulatory system; effects on certain antigens, as well as the stimulation of specific antibodies, by increasing their affinity for bacterial and viral proteins toxic bacterial components;
- effects on the coagulation process, which account for the hemostatic action.

All these applications are considered to be based on the saline effect of NaCl as a strong electrolyte that can affect, at certain levels of concentration, the epithelial system and the microbial flora as well as the humoral system and the superficial nerves (Sandu et al., 2009a; 2010a). The NaCl nanostructures may take three forms: as dry glomerulate microparticles (dry aerosols); as hydrated microparticles (hydrated solvions or aerosols with a monomolecular layer of water); as nanostructures dispersed in aqueous systems, going as far as simple hydrated ions ($Na^{+}_{(aq)}$ and $Cl^{-}_{(aq)}$).

The glomerulate (grouped in small, dense clusters) structure, which depends on the degree of hydration, can take shapes that resemble snowflakes.

Table 1. The dimensional groups and the forms of the NaCl aerosols

Type of Particle	Ions	Aitken	Average-size	Large	Huge
Form	Small simple ions	Large ions and small ionic aggregates	Condensate particles	Coagulated particles	Sedimentable particles
Radius of particle (nm)	< 0.5	0.5...50	50...100	100...1000	> 1000

The shape and size of such nanoparticles is determined by a series of environmental factors, mainly by the ones depending on the acid-basic reactivity of the environment. The nanodispersions of the solvions in the biological extra-cellular fluids can reach a concentration of cca. 0,9% NaCl, whereas in the intra-cellular fluids the concentration is lower due to the specialized membrane channels that allow the direct and/or the reverse osmosis of the $Na^+_{(aq)}$ cations which, comparatively, have ionic volumes larger than those of the $Cl^-_{(aq)}$ anions.

The nanoparticles dispersible in the air behave as ”negative aeroions”, as they are glomerulate nanocrystalites, either dry or hydrated, with negative surface charges. Such negative solvions have beneficent effects on organisms. Moreover, in the air they interact with organic nanodispersions, either dry or with one-layer hydration, which behave as ”positive aeroions”, such as the ones resulting from pyrolysis, cracking, or burning of cigarettes, in which cases the toxic effect of smoke is eliminated by the electrostatic destabilization of the dispersion (Alfoldy et al., 2002; Sandu et al., 2009a; 2010a).

Ethnographic surveys mention the fact that most of the cataplasms used in treatments, such as pouches with salt and even thin strata of salt, should be warmed to an optimal temperature for the human body (around 50-60 degrees Celsius). Usually, the users do not think about the thermal effect. It is true that the thermal shock produces certain improvements, but only during the application. Therefore, the curative effect is mostly due to the aeroions emanated by the salt and inhaled into the lungs and thus acting upon the epithelia through osmosis. It also activates the immune systems, including the leukocytes; it stimulates the formation of antibodies and it improves the potentially pathogenic biochemical processes. In this sense, the nanostructural NaCl aeroions play the multiple role of an antigen at the level of certain organs, producing most of the previously mentioned effects (Chervinskaya, 2007; Hedman et al., 2006; Poryadin et al., 2002; Sandu et al., 2002; 2003; 2006; 2009a; 2010a).

Another therapeutic application that needs to be explained is the use of the fine dispersions of salt having as basis vinegar, wine and oil. We know that the solubility and the formation of certain crystalline solvates and of certain hydrolyzable molecular species formed in situ in a dispersed system depend on the pH. They also depend on the presence of ions other than sodium and chloride, on temperature and on other factors that favor the formation of certain nanostructural solvated aggregates of NaCl. In the latter the superficial single layer of water has a dipolar profile with external negative charge, which favors a behavior similar to the one of the dry negative aeroions that stimulate the recovery from certain diseases by biochemical mechanisms specific to the inorganic antigens. Vegetable oil as well as wine and vinegar create a mildly acidic pH and favor the specific nanostructuring of the crystallites with superficial negative charge, capable of osmotic processes through the membranous systems of the internal organs. Also, the membranous systems based on polypeptide structures with reactive support have an influence on the re-formation of the NaCl structures of a solvion type, due to the fact that such systems contain amphionic groups. As a result of the electrostatic interactions, the polypeptides enhance, by their isoelectric point, the ordering of flexible and mobile structures. Thus, functional biotic microstructures of certain tissues are optimized (Sandu et al. 2009g; 2010). Several concrete applications are known in cases such as the lumbago, the stomach, the liver and the pancreas aches. These applications are added, in combination with vinegar and wine, to the sodium chloride and to the small amounts of bronze or copper filings, and are consumed after decanting. Nevertheless, we do not have enough explanations for such treatments (Poryadin et al., 2002). What we may observe here is that the copper filings can be anodically dissolved ($Cu^0 \rightarrow Cu^{2+} + 2e^-$) in the presence of the NaCl electrolyte, in a solvion form ($Cu^{2+}_{(aq)}$) that interacts with the wine tannins in an acid medium, the result being molecular species that are hard to hydrolyze or dissolve. Copper filings provide completion of the macroelement deficit in certain enzymatic processes. Also, in the presence of the NaCl as a highly effective electrolyte, copper filings prove to play an important part in the elimination of toxicity from the red tannin-rich mountain wines (Sandu et al., 2009a).

More recently, NaCl with KCl and other salts are used in artificial halochambers, for terapy and in order to improve the performance of sportsmen. Thus, the KCl aeroions are fundamentally involved in a massive amount of body processes, such as fluid balance, protein synthesis, nerve conduction, energy production, muscle contraction (conversion of glucose into glycogen), synthesis of nucleic acids and control of heartbeat, reducing the blood pressure. In many of its roles, potassium is opposed by sodium, and the two positive ions are jointly balanced by the negative ion, chloride (He et al., 2005; Matsui et al., 2006; Pikilidou et al., 2007).

4. Present-day procedures for producing the saline aerosols

Besides the halochambers from the salt mines, very much used both for prophylactic purposes and in the therapy of some diseases, there have been drafted after 1990 a series of procedures for obtaining the saline aerosols on the basis of NaCl as such or in mixture with some inorganic or organic compounds, with pre-established chemical compositions depending on the application type. Depending on the production physico-mechanical, hydric and thermal processes, these procedures are classified into four groups; here they are:

- *mechanical separation* or *erosion*, followed by *physical dispersion* in the atmosphere from the halochamber with the help of a gas flow, of the saline systems under the form of esorated precipitates, fine micro-crystallites, extruded micro-pellets or pellets obtained through recrystallization from supersaturated solutions by means of the hydro-thermal processes or by the evaporation of the solvent from the thin layers of concentrated solutions accomplished by dripping (Clark et al., 1996; Hickey, 1994; Pascu, 2002a; 2002b; 2003a; 2003b; 2003c; 2006; 2007; 2008; 2009a);
- *breaking of the gas bubbles* in the barbotage processes with air or other inert gases by means of the supersaturated saline solutions (Joutsensaari et al., 2001);
- *atomization of the saturated saline solutions* in vacuum centrifugal air separators, followed by the physical dispersion with the help of an air flow (Katusik et al., 2000; Merchat, 1994);
- *carrying-away of the superficial particles* resulted from solvolyses and the consecutive anhydrizations of the surface structures as a result of the air carrying through the holes and grooves done in the bars or parallelipipedic blocks of halite, arranged as screens on the walls of the halochamber (Albiach, 1995; Belkin, 2005; Konovalov, 1993; Sandu et al., 2009b; 2009c; 2009d; 20092; 2010b; 2010c).

In the case of the first group of methods, the discharge, the capacity and the output of the source (installation, device or apparatus) depend on a series of factors such as:

- the *degree of separation* of the micro-crystallites,
- the *intensity of the gas flow*,
- the *complexity of the installation* and the *geometric section* of the active zone (filters, frames, honeycomb-type structures or diaphragms, platforms, cross walls),
- the *humidity* and the *temperature of the carrying gaseous agent*, the *temperature* and the *humidity* at which the dispersed saline material is being kept,
- the *operating conditions of the installation* (the ratio between the operating period and the non-operating one) etc.

In the case of the large crystals, the processes of *deliquescence* and *efflorescence* which take place during the non-operating periods improve the discharge or the capacity of the installation.

With this end in view one knows the SALIN devices manufactured by the TehnoBionic Company, Buzău (Pascu, 2003a, 2007; 2008; 2009a), devices which, by means of ventilation, carry out the particles resulted from *solvolyses* and the *consecutive anhydrizations* from the surface of the pellets obtained by recrystallization from the supersaturated solutions of NaCl as such or in the mixture with others salts (KCl, $MgCl_2$, $CaCl_2$, KI, etc.).

In what the second group of methods is concerned, there are four stages (Fig. 1) for producing the NaCl aerosol particles (Moore et al., 1954; Pruppacher and Klett, 1978). The chemical composition of the aerosol particles depends on the components existing in the supersaturated saline solutions while the micro-physical and nano-structural characteristics depend on the temperature of the saline solution and of the bubble-generating agent, on the pressure of the gas as well as on the size of the gas bubbles. For instance, one knows that the number of the aerosol particles produced by the breaking of the gas bubbles increases with the increase of their sizes. A bubble with a size of some millimetres forms, by breaking, some hundreds of particles, and the average frequency of their forming is 25 - 100 particles·cm^{-2}·s^{-1} for a slight gas sparging (Blanchard, 1969).

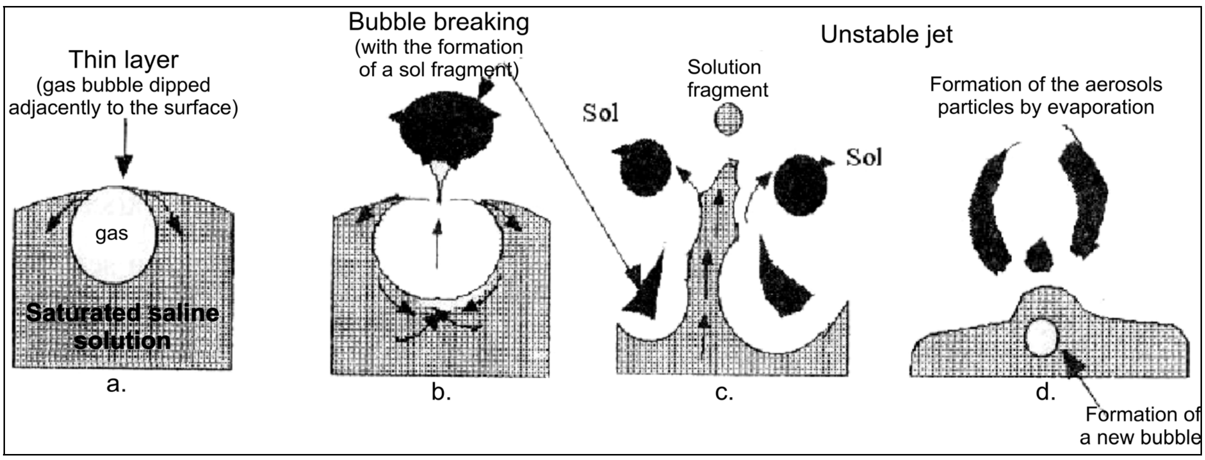

Fig. 1. Stages for the production of the saline aerosol particles in the barbotage system by means of the mechanism of the gas bubbles explosion: a. Formation of the thin layer at the surface of the saline solution (gas bubble dipped at the surface); b. Bubble breaking with the formation of a solution fragment; c. The unstable jet produces the breaking and the formation of some other fragments from the adjacent layers; d. Formation of the fine particles after the evaporation of the solvent from the fragments of the solution dispersed in the siccativant gaseous medium

With the third group of methods, depending on the diameter of the spray nozzle, correlated with the discharge of the jet as well as with the value of the vacuum pressure and temperature, one can obtain particles with dimensional distributions preferential for one of the four dimensional groups. By comparison with the first two groups of methods, the last one has the disadvantage both of using some complex equipment and of high production price.

The fourth procedure group which implies the *carrying-away of the superficial particles* resulted from the consecutive processes of solvolyses and anhydrization of the superficial structures from the holes and grooves done in the parallelepipedic blocks of halite, arranged as screens on the walls of the halochamber, taking over to a certain extent the structure and the functionality of the neutral system of the halochambers from the old salt mines. With this end in view there exist a series of artificial salt mines in which the particles from the surface of the salt blocks are taken over by conveying the air by means either of the conventional jets or of an air jet from a blower (Albiach, 1995; Anderson, 1995; Belkin, 2005; Clark et al.,1996; Gafurov, 1990; Hickey, 1994; Kabanov, 1992; Klaus, 1993; Konovalov, 1993; Sandu, 2009a; 2009b; 2009c; 2009d; 2009e; 2009f; Wills, 1995).

5. Conclusions

This paper represents the first report from a series which includes the state-of-the-art analysis and synthesis concerning the structure, the form and the sizes of the particles of NaCl *aeroions* in correlation with the production methods and their practical applications.

Depending on their *activity* and *life cycle*, respectively, the aerosol particles have a somewhat steady dimensional distribution, which is the result between the *production rate* and the *loss* during different processes (condensation, coagulation, electro-neutralization, sedimentation/destabilization etc.). From this point of view, these particles are classified in five groups (*simple ions, Aitken particles, small particles*, *average-size particles, large* and *huge particles*).

According to the historical sources and the present-day literature, the salt has multiple applications either as brine or marsh, under a solid form or as aerosols, being used for the treatment of various diseases, in the air purification and for increasing the human performances.

Historical data rendering evidences the presence of an ethnoscience, which has allowed the subsequent development of the modern biochemistry and bio-physics, thus substantiating some actual applications, the emphasis being put on halotherapy.

Regarding the production of the aerosols, this paper presents the four procedure groups, namely, the separation, the breaking of the gas bubbles, the atomization of the saturated saline solutions, and the carrying-away of the superficial particles from the salt blocks.

In what the characteristics of the aerosol particles are concerned, these are determined not only by the source but also by the environmental factors. Accordingly, there are discussed the functional characteristics, specific to a source of saline aerosol (the *size and density of the aerosol*, the *forming rate of the particles*, the *source output*, the *factor for the enrichment of the gaseous medium*, the *life cycle of the particles*). All these are used to choose the optimal generator model and the nano-structural and micro-physical properties of the aerosols (the *aerosol concentration* and its *variation in time*, the *dimensional distribution* of the particles, the *dynamic behaviour* of the aerosol, the *diffusion*, *mobility* and the *drift speed* of the particles as well as the *limit humidity* of the environment at which the formation of the condensation nuclei starts). These lead to a proper understanding of their implication in the climatic environments.

There are presented the three specific areas of a climatic environment activated by a source, namely, the *active layer* (the area near the source), characterized by high concentration where all four dimensional groups of particles coexist; the *diffused layer*, an extended one, characterized by a *dynamic state* of the *distribution of the dimensional domain* and of the *life cycle*, having all parameters in a continuous variation; the *residual* or *passive layer* (the most remote from the source or existing in some hardly accessible zones) with a small and usual uniform concentration, characterized by a *stationary state* of the *distribution of the dimensional domain* and of the *life cycle*.

The dominant processes in these three zones are: the *condensation* based on chemical reactions and which form particles from the *nucleation group*, the *condensation* or the *peptization* of the primary particles, thus resulting the *accumulation group*, and, finally, the *coagulation* and *monolitization* of the large and huge particles, thus resulting the group of the *sedimentable* particles. Among these, the greatest importance in the establishment of the climatic environments with a therapeutical and ambiental purpose is awarded to the group of the accumulation particles that, in fact, form the *diffused layer* as well as by the group of the very small particles from the *active layer*.

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