ЗАГАЛЬНІ ПРОБЛЕМИ ЕКОЛОГІЧНОЇ БЕЗПЕКИ

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ATMOSPHERIC AEROSOL ECOLOGICAL STATE REMOTE SOUNDING BY SPECTROPOLARIMETERS

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Atmosphere pollution leads to negative ecological and climate consequences, causes diseases and mortalities. Therefore it is important to develop and introduce new effective technical means of global monitoring of aerosol state and dynamics in the Earth atmosphere. In the present paper improvement of atmospheric aerosol ecological monitoring system by implementation of satellite small-size spectropolarimeters capable of measuring 4 Stokes parameters is proposed. This enables obtaining data on atmospheric aerosol optical density, mean refraction and absorption indexes, maximum and dispersion of size distribution function. *Key words:* ecology, monitoring, atmosphere pollution, atmospheric aerosol, spectropolarimetry, Stokes parameters.

Дистанційний моніторинг екологічного стану атмосферного аерозолю за допомогою спектрополяриметрів. Патлашенко Ж.І. Забруднення атмосфери призводить до негативних екологічних і кліматичних наслідків, спричинює захворювання та смертельні випадки. Це зумовлює актуальність розроблення та впровадження нових ефективних технічних засобів глобального моніторингу стану й динаміки аерозолю в атмосфері Землі. У роботі запропоновано вдосконалення системи екологічного моніторингу атмосферного аерозолю за допомогою космічних малогабаритних спектрополяриметрів, здатних визначати чотири параметри Стокса. Це дасть змогу отримувати дані щодо оптичної густини атмосферного аерозолю, середніх значень показників заломлення й поглинання аерозольних частинок, максимуму та дисперсії функції розподілу частинок за розмірами. *Ключові слова:* екологія, моніторинг, забруднення атмосфери, атмосферний аерозоль, спектрополяриметрія, параметри Стокса.

Дистанционный мониторинг экологического состояния атмосферного аэрозоля с помощью спектрополяриметров. Патлашенко Ж.И. Загрязнение атмосферы приводит к негативным экологическим и климатическим последствиям, вызывает заболевания и смертельные случаи. Это обуславливает актуальность разработки и внедрения новых эффективных технических средств глобального мониторинга состояния и динамики аэрозоля в атмосфере Земли. В работе предложено усовершенствование системы экологического мониторинга атмосферного аэрозоля с помощью космических малогабаритных спектрополяриметров, способных определять четыре параметра Стокса. Это позволит получать данные относительно оптической плотности атмосферного аэрозоля, средних значений показателей преломления и поглощения аэрозольных частиц, максимума и дисперсии функции распределения частиц по размерам. *Ключевые слова:* экология, мониторинг, загрязнение атмосферы, атмосферный аэрозоль, спектрополяриметрия, параметры Стокса.

Introduction. Light scattering in the Earth atmosphere is a complex process that depends on atmospheric environment state and atmospheric components concentrations, first of all on temperature and pressure of the atmosphere at certain altitudes [1].

Atmospheric aerosol ecological and climate state of the atmosphere. Aerosol particles sized from a few nanometers to a few micrometers have diverse compound, structure and physical properties [2–5]. Despite the complexity of aerosol interaction with light it's optical properties are approximated by seasonal/regional models used as a basis for remote optical investigation methods. Such approach provides for consistent results in case of standard atmospheric state. But in case of non-regular natural or technogenic aerosol emissions (volcano eruptions, hurricanes, sand storms, anthropogenic point and surface sources, etc.), most of contemporary models do not provide for reliable quality of data interpretation, leading to impossibility of aerosol ecological impact determination. This problem is exceptionally important in case of global ecological monitoring, when monitoring data is interpreted based on high-scale models that do not consider local peculiarities and is often not adjusted to ground monitoring sites data.

Aerosol environmental impact. Aerosols form due to different physical processes, such as evaporation and

condensation, turbulent gas and hydrodynamic processes, photochemical and chemical reactions, etc. [6]. Aerosol also forms a result of anthropogenic activities like manufacturing, mining, building industries, burning of oil products, coal, gas, etc. The aerosol may also be returned in the atmosphere once sedimented lifted by wind or evaporated. The largest aerosol quantity is sea aerosol, and anthropogenic aerosol constitutes around 10% of total aerosol mass [7].

Atmospheric aerosol influences global climate by changing the Earth albedo, and by changing transparency of Earth atmosphere in infrared. These phenomenon are described as a set of direct, indirect and semi-indirect aerosol effects [8–11].

Direct aerosol effect is caused by absorption and scattering of light by atmospheric aerosol. Indirect aerosol effect is caused by Earth albedo change due to altering optical and physical clouds characteristics. Aerosol as condensation nuclei increases raindrops number (first indirect effect or Twomey effect [9, p. 12–14]) and volume, delaying rainfall and prolonging the cloud lifetime (second indirect effect or Albrecht effect [10, p. 13–15]).

Semi-direct aerosol effect includes all effects that do not fit into direct and indirect aerosol effects definition, such as moisture height redistribution [16], vertical moisture fluxes stabilization [17], vertical temperature profile change [10], etc.

An important ecological phenomenon is atmospheric aerosol deposition that changes Earth surface albedo and ecological state. An example of this effect is anthropogenic aerosol deposition on Arctic and Antarctic ice that causes its melting due to albedo decrease, estimated at 10% [18].

Aerosol directly influences the environment and eventually directly or indirectly affects humans. Since 1970s many mortalities are linked to anthropogenic atmosphere aerosol pollution [19–21]. Yearly death rate in USA linked to atmospheric aerosol is estimated to be 22 to 52 thousands [22]. And total quantity of respiratory diseases due to aerosol pollution is reported to increase by 50% every 5 years [23].

It is well known that inhalation of small aerosol particles may cause asthma, lungs cancer, cardiovascular diseases, respiratory diseases, birth defects and premature death. Large aerosol particles are mostly filtered by epipharynx. However, particles with size under 10 micrometers may be transported deeply into lungs – to bronchi and alveoli. These particles are regulated by PM10 standard in EU [24].

Particles under 2.5 micrometers are regulated by PM2.5 EU ecological standard and may be transported to lungs gas exchange areas causing plaques in arteries, inflammation of blood vessels, arteriosclerosis, and other cardiovascular diseases [25; 26]. According to World Health Organization estimates PM2.5 particles cause around 3% mortality due to cardiopulmonary diseases, 5% mortality due to trachea, bronchi and lungs cancer and 1% mortality due to acute respiratory infection in children under 5 years old [19]. Moreover, such diseases may emerge due to short-term inhalation of corresponding aerosol particles [27].

Aerosol particles under 100 nanometers (e. g. exhausts of so-called clean diesel engines) freely pass through lungs to blood and directly disorder internal organs including brain. This aerosol can carry carcinogenic compounds such as benzopyrenes.

Aerosol particles permeability to human organs is determined not only by their size, but also by their shape and physical-and-chemical compound [28]. The problem of atmospheric aerosol ecological risk dependency on aerosol shape is underexplored. Only general considerations that "sharp edged" aerosol particles (like asbestos) are more dangerous than "smooth edged" particles are formulated. Nanoscale aerosol particles that have increased surface area (i. e. irregular shape) in comparison to spherical particles have higher chance of accumulating different hazardous substances at their surface.

Total mass of atmospheric aerosol contamination is not directly related to its ecological hazard, e. g. a single 10 micron particle is more environmentally safe than a thousand of 100 nm particles that have 100 times less total mass. In some countries a total aerosol surface area regulation is proposed.

Another source of ecological hazard of natural and anthropogenic aerosol pollution is indirect impact at human health through food products of animal and vegetation origin, and through damage of natural ecosystems. E. g. increased atmospheric aerosol concentration may cause death of some plants [29].

Moreover, aerosol may pose direct hazard to health of living organisms, e. g. highly toxic aerosol or bacteria and viruses. Radioactive atmospheric aerosol represented by 0,02 to 1 micron particles, e. g. formed due to nuclear accidents similar to Chernobyl and Fukushima-Daiichi, creates a high ecological risk. Such radioactive aerosols are referenced to as "low-activity" (radioactivity levels lower than 10-13 Ci), "semi-hot" (radioactivity levels range from 10-13 to 10-10 Ci) and "hot" (radioactivity levels higher than 10-10 Ci). According to the origin they are separated into natural, explosive (formed in nuclear device detonation) and industrial (formed during nuclear substances management).

Radioactive aerosol inhalation or consumption is much more hazardous for living organisms than equivalent external irradiation as they can sediment in the body and create internal irradiation which directly influences internal organs by focal necrosis. Only 10% to 50% of radioactive aerosols can be efficiently removed from the body. Average radioactive aerosol troposphere suspension time varies from 2 to 30 days depending on its origin and local weather and may be quickly transported around the globe. E. g. I131 aerosol from Fukushima-1 nuclear disaster was registered in Ukraine in just 16 days after the accident.

Contemporary state of atmospheric aerosol monitoring. Constant monitoring of atmosphere aerosol con-

tamination is performed in many world countries and regions. All atmospheric aerosol monitoring methods are separated in two distinctive classes: contact probing and remote sounding.

Local survey is made near the point pollutant objects [30], around perimeter of area pollution sources [31] and near population centers or special ecological control areas.

Contact probing is made by special hydrometeorological laboratories by analyzing chemical compound of atmospheric precipitation or by atmospheric air sampling and include aerosol mass spectrometry, differential mobility analysis, aerodynamic particle sizing, wide range particle spectrometry, micro-orifice uniform deposit impactors, condensation particle counters, epiphaniometry, electronic microscopy, instrumental neutron-activation analysis, etc.

On one hand contact direct sampling is the most accurate and reliable atmospheric aerosol ecological state investigation method, that provides for their implementation in international and national law. On the other hand such methods have to be performed on-site and therefore are confined spatially.

Non-contact remote sounding methods provide for lower accuracy and reliability in comparison to contact probing laboratory analysis. However they enable implementation of cost-efficient atmospheric aerosol parameters estimate. Classical remote sounding methods include solar photometers, polarimeters and LIDARs that provide for aerosol optical density determination. In some cases aerosol particles size distribution function maximum can also be determined.

Remote sounding methods are separated into active and passive. Active methods include LIDARs investigating laser emission scattering by atmosphere. It provides for aerosol vertical distribution estimation and does not depend on phase angle. However, such method requires rather complex infrastructure.

Indirect sounding methods also include monitoring specific aerosol type influence at different atmospheric components or underlying surface. A good example is air ionization by radioactive aerosols detection by standard military or civil radars providing for quantitative estimate of its emission and transfer.

Passive methods investigate solar light scattered by Earth atmosphere. Therefore such methods are influenced by additional uncertainty linked to solar radiation parameters fluctuations. Phase angle temporal variation and different atmospheric components spectra overlap due to rescattering, luminescence, reabsorption of solar light, etc. introduce additional biases. These problems can be solved by direct solar calibration [32], most efficient in space conditions, and by multifrequency spectrometry or spectrophotometry.

Remote sounding methods may be ground or airplane/satellite based. To solve the problem of satellite data interpretation ambiguousness the ground stations are used as validation for global satellite monitoring [33].

As a consequence, current state of atmospheric aerosol monitoring does not solve the problem of high-quality global ecological monitoring completely: local ground-based contact and remote investigation stations despite their accuracy are limited spatially and satellite methods lack accuracy and reliability and are limited by quantity of measured parameters.

A quality step in optical remote sounding methods is simultaneous detection of spectral and polarization parameters (Stokes parameters) of the optical signal that would increase the amount of the experimental data obtained 4 times.

Problems of atmospheric aerosol scattering matrix remote monitoring. Atmospheric aerosol particles interaction with light is significantly different from gas atoms and molecules which differs first of all for polarization properties of the scattered light.

The atmospheric aerosol scattering matrix is a quasi-symmetric 4×4 Muller matrix that describes transformation of incident light Stokes vector to scattered light Stokes vector [34]. Specific values of scattering matrix components depend on averaged atmospheric aerosol characteristics, their dispersion and on predominant orientation of anisotropic particles relative to incident light. In some cases the matrix form is simplified by particles shape symmetry.

All 16 independent scattering matrix components can be derived only in case there is a way to control the incident light polarization, i.e. by using active Stokes-polarimetry. And determination of specific aerosol properties inverse problem may be accurately solved only in case scattering matrix components are known for a wide set of phase angles. Moreover, experimental errors will cause such inverse problem to be an ill-posed problem and therefore will require regularization. Active Stokes-polarimetry may be implemented at autonomous probes in the Earth atmosphere [35], however, limitation of such methods are the same as for contact probing.

LIDARs provide for both incident light polarization control and vertical distribution determination. On the other hand, LIDARs have their engineer and technical limitations both from size and mass and from power consumption point of view. Moreover, reverse scattered light by most atmospheric aerosol types has nearly identical polarization to that of incident light, therefore a synchronous observation system must be established of spatially separated lasers and detectors, limiting system mobility and efficiency (e. g. synchronous ground-and-space systems are also possible e.g. to validate satellite monitoring data). Further, even in case of a limited amount of spatially-separated detectors is present, the polarization is still determined at a discrete set of phase angles. And finally, there are ecological problems in operating wavelength-tunable lasers that contain environmentally hazardous organic dyes and acids.

On the other hand passive devices can be relatively compact to be used in a space/aerial vehicle. At present only spectrometers (measuring only 1 Stokes param-

eters) and photopolarimeters (measuring 2 and more Stokes parameters in a single wide spectral band) are used without providing for ability to separate gas and aerosol spectra in the atmosphere and to cancel out underlying Earth surface spectral albedo.

Therefore there is no efficient contemporary method of atmospheric aerosol scattering matrix monitoring.

Aerosol light scattering peculiarities. Aerosol light scattering is usually separated in two different cases: small particles (particle size smaller than the wavelength) and large particles, which are usually considered spherical. However, spherical particles are not observed in nature and even a raindrop has aspherical shape due to gravitation forcing and gas dynamics. Crystals, snowflakes, smoke particles and dust formed by minerals fragmentation surface shape is very complex and scattering matrix model determination is a complex theoretical problem [36]. Rough particles are a separate class of aerosols, which have surface inhomogeneities size comparative to the wavelength [37]. The diversity of forms and structure of polydisperse aerosol ensemble is hard-to-model and may significantly influence monitoring data interpretation.

It was shown [38] that spherical atmospheric aerosol model (Mie theory) gives incorrect results for chaotic oriented fractured glass particles and phase dependencies of matrix components are not consistent with experimental data. Significant discrepancies in linear polarization degree of crystal ammonia formed at temperatures from 130°K to 180°K at wavelengths 470, 652 and 937 nm were also found experimentally [39].

Main discrepancies between spherical and chaotic oriented aspherical particles are following: spherical particles have more intense interference structures in second and third Stokes parameters and scattering indicatrix phase dependencies and higher reverse scattering intensity. However, total single-scattering albedo (first Stokes parameter) does not significantly depend on particles shape peculiarities [40].

Second and third Stokes parameters value and indicatrix for large particles strongly depends on refraction index and size distribution function parameters. Frequency and amplitude of the polarization degree oscillations relative to phase angle, particle size and refraction index are larger for monodisperse aerosol and smaller for polydisperse aerosol. Moreover, direct and reverse scattering intensity increases with increase of size dispersion, while reverse scattered light intensity is proportional to refraction index.

It should also be noted that the aerosol size is defined only for spherical particles. In case of aspherical particles “equivalent volume” (identical volume sphere) and “spherical shape factor” (sphere to particle surface area ratio) are introduced. Sometimes “sedimentary” radius is also determined being the sphere radius with equal sedimentation speed.

Off-atmospheric aerosol remote spectropolarimetric monitoring. Phase dependency of second and third Stokes parameters determination (fig. 1) is one of the most promising methods for refraction index determination.

Assuming that polarization degree is formed at high altitudes [36], atmosphere vertical inhomogeneity and particles vertical stratification may be neglected. In case absorption index is less than 10-3 (which is true for most natural atmospheric aerosols), the second and third Stokes parameters are determined mainly by refraction index.

Second and third Stokes parameters analysis in a wide set of phase angles provides for the most accu-

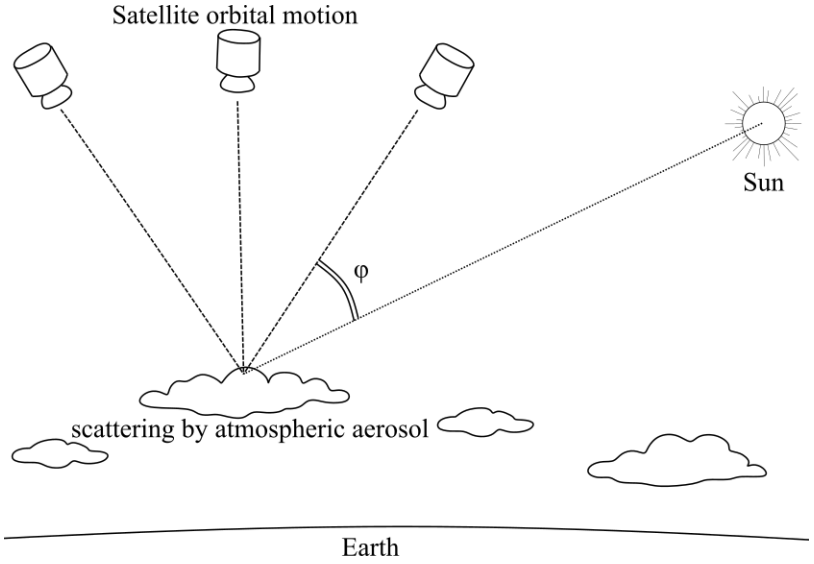

Fig. 1. Atmospheric aerosol monitoring geometry, φ – observation phase angle, i. e. the angle of the light ray from the source to the observed volume and the light ray scattered in the observer direction

rate monitoring data and enables simultaneous estimation of refraction index, maximum and dispersion of size distribution function, and optical absorbency by comparison with the model. However, practically their spectral dependency may be measured instead under assumption that refraction index dispersion is negligibly small or known, which is approximately true for many types of atmospheric aerosols of different origin in visible light.

There is also a method for atmospheric aerosol parameters determination by measuring fourth Stokes parameter [41], which is determined only by multiple scattering. The obtained aerosol parameters are averaged by a larger atmosphere depth and can be different than that obtained by second Stokes parameter measurement. Therefore the difference carries information on aerosol vertical distribution.

Absorption index can be estimated by aerosol single-scattering albedo spectral dependency [42]. However, such method has a high error due to neglecting aerosol vertical distribution and strong dependency on aerosol particles shape model [43].

Conclusions. The atmospheric aerosol ecologic parameters may be determined by analyzing scattering matrix. Given some preliminary knowledge on aerosol nature and particle shape the scattering matrix may be determined through one-channel observations at wide range of phase angles or multichannel observations at a limited range of phase angles. The first approach has higher accuracy, while the second one provides for highly dynamic processes investigation and global monitoring implementation.

Second and third Stokes parameters of backscattered light in the atmosphere spectral and phase dependencies enable estimation of atmospheric aerosol averaged refraction and absorption indexes, maximum and dispersion of size distribution function. Fourth Stokes parameter enables validation of the obtained data.

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