Aerosol formation in the filtered atmospheric air and regularities of their evolution

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Abstract. Presented is the study of atmospheric aerosol particle formation and evolution processes arising in filtered atmospheric air in complete darkness. Studies were carried out in the Large Aerosol Chamber (LAC) of RPA «Typhoon» with 3200 m³ volume under conditions close to natural atmospheric ones. The large volume of the chamber and its design features make it possible to exclude the influence of boundary conditions as well as any additional pollution sources from the chamber walls and from the equipment inside the LAC on the processes under study. We observe the formation of nanoparticles apparently due to gas-to-particle conversion in the absence of UV radiation. We investigate these newly formed particle growth regularities from 15 nm size to cloud condensation nuclei characteristic sizes under controlled conditions. The observed evolution regularities of the newly formed due to gas-to-particle conversion aerosol spectra can significantly contribute to the understanding of atmospheric aerosol formation processes responsible for cloudiness and precipitation.

1. Introduction

The atmospheric aerosol is among main elements that significantly impact the climatic state of our planet [1]. The formation of cloud systems, which are the most important part of the evaporated water recuperation from seas and oceans back to lands, looks impossible without atmospheric aerosols. The investigation of atmospheric aerosol formation, evolution, and destruction is among actual problems for planetary climate models.

Today, after basic works of [2], it is considered [3] that hygroscopically active atmospheric aerosols are formed by nucleation of the so-called conversion gases in atmospheric air. These particles further enlarge its size up to a median count diameter of 100 – 300 nm due to Brownian coagulation and can serve as suppliers of condensation nuclei for the nucleation of cloud droplets.

It is important to register newly formed aerosol particles and its evolution in the outdoor air under conditions as close as possible to those typical for the natural atmospheric ones with minimal influence of boundary conditions. Available at RPA «Typhoon», the Large Aerosol Chamber (LAC) with 3200 m³ volume is equipped with two (external and internal) HEPA 13 aerosol filters [4]. LAC design features allow a complete modeling of the processes occurring in the natural atmosphere. The LAC allows achieving practically zero aerosol particles concentration due to filters for cleaning outdoor and indoor air from aerosols. Moreover, because of the experiment demand, after secondary purification in the LAC, the achieved state with a low (practically zero) aerosol concentration is always maintained for an indefinitely long time (up to 10 days or more). It means that chamber walls and equipment installed inside the chamber do not produce aerosol particles. We can avoid a significant effect influenced by the walls on the
deposition of the aerosol due to the chamber large volume. This opens the possibility of studying various processes of the formation and evolution of atmospheric aerosol in the LAC under close to natural conditions.

2. Registration of aerosol generated in the large aerosol chamber (LAC) isolated from the external environment

We carry out an experimental study of aerosol particle formed from atmospheric air gas component in the chamber. The LAC is a sealed steel cylinder of 15 m diameter and 18 m height. The detailed description of its design and thermodynamic characteristics is described in [5]. Air filters HEPA 13 (High Efficiency Particulate Air) are installed into the LAC. The filters purify the air to almost zero aerosol particle concentration in whole size particle range of the devices. The first filter is in the channel for filling the chamber with atmospheric air (the external one), while the second filter is inside the LAC (the internal one). The Model 3936L88-N of Scanning Mobility Particle Sizer SMPS by TSI is designed for the size distribution of aerosols from 15 nm to 1000 nm. The spectrometer measures the size distribution of particles using an electrical mobility detection technique. The system has 115 measurement channels. We take air samples from the LAC through a galvanized steel tube with the internal diameter of 18 mm and the length of 2 m. The tube passes through the chamber wall.

Figure 1 shows the experimental results. Let us divide the experiment into several stages. The purified from aerosol outdoor air fills the LAC on the first stage. We draw this stage in Figure 1 with the line from time $t_0$ to time $t_1$. Before time $t_0$ the particle number concentration corresponds to the particle number concentration in the outdoor air. The air in the LAC was purified by pumping outdoor air through an external filter (the purified level is up to several tens of particles per cm$^3$).

![Figure 1. Time dependence of the particle number concentration C in LAC in the process of aerosol evolution after filling with outdoor air passed through the external and then internal filters.](image)

On the second stage, which one can see in Figure 1 from time $t_1$ to time $t_2$, we register aerosols in the LAC. The figure shows that the particle number concentration in the LAC on this stage in some moments is almost twice above the background concentration. So, the process of new particle formation and evolution probably takes place in LAC on the second stage of the experiment.

Then we again purify the air from aerosols in the LAC by using the internal filter. This third stage lasts of about two hours. Figure 1 shows this stage by the line from time $t_2$ to time $t_3$. The particle number concentration in this stage decreases and becomes $C \approx (10 – 20)$ cm$^{-3}$. 
Then, in the last fourth stage, the particle concentration in LAC does not increase and practically does not change during 300 hours.

We carry out a series of such experiments. The experimental results show that after secondary filtration the particle number concentration always becomes almost zero. This low concentration stays for a long time (up to 10 days and more).

3. The studies of regularities of newly-formed aerosol evolution

As pointed above, the second stage can be characterized by the registration of the increasing aerosol concentration in the LAC. This testifies that in purified air inside the LAC, in full darkness, we probably observe the newly-formed aerosol creation. These results approve the known opinion [1-3, 6], that there is the natural process of aerosol formation, but nobody knows the mechanism of the process [7].

Figure 2 shows the temporal evolution of aerosol spectra. The curve with index «ext» presents external air aerosol spectra with concentration \( C_{ext} \approx 4 \times 10^3 \, \text{cm}^{-3} \). Index «0» corresponds to the beginning of the aerosol spectra measurement in the LAC, which coincides with the end of filling the LAC with purified air (the air concentration \( C_0 = 27 \, \text{cm}^{-3} \)). Insofar as in this time moment, we register particles only by one channel, the curve «0» is the separate markable points. Other indexes are the corresponding measurements started from this time moment in hours.

![Figure 2. Secondary aerosol formation and evolution in LAC after filling it and purifying the air in it with an external H13 filter (30/08/2018).](image)

Curve numbers correspond to the 10-minutes cycle measurement starts (in hours) from the moment of the end filling. The vertical line denotes the lower SMPS threshold.

One can see from Figure 2 that after 1/3-hour, an additional aerosol with a size above the threshold in 15 nm appears by the residual aerosol background. The complete distribution function forms after three hours. The full aerosol spectra forms after 20 hours. As shown in figure 2, no particles are less than 20 nm in the curve with the «127» index. Thus, we can conclude that the aerosol lifetime for a particle with a size below 20 nm is less than 20 hours.

Figure 3 gives the time dependence of the integrals of the spectra shown in figure 2.
Figure 3 – Time dependence a) median diameter $d_m = d_{10}$, b) relative breadth $rb$ and relative asymmetry $ras/10$

Although median diameter of particle changes from 30 to 73 nm during evolution time from 3 to 147 hours, as shown in Figure 3, relative breadth $rb$ and relative asymmetry $ras$ vary insignificantly – these parameters ($rb$ and $ras$) describe regularities of the distribution of the particles by their sizes. The relative breadth value asymptotically tends to 0.27 – 0.29. The relative asymmetry is positive and hesitates near the value of $ras \approx 2$. Based on this we choose the gamma distribution as the most appropriate approximation for spectra evolution. From Figure 4 one can see that the approximation is quite well.

Figure 4. The experimental (with markers) and approximated theoretical (without marker) newly formed aerosol spectra for time evolution moments 92, 117 and 147 hours.

4. Conclusion
The Large Aerosol Chamber (LAC) RPA «Typhoon» of 3200 m$^3$ volume allows conducting the research of gas-to-particle conversion process under conditions close to the natural atmospheric ones. The newly-formed particle creation process we observe in the complete darkness and under the absence of UV radiation.
After removing newly-formed aerosols, we do not observe new particles for a long period (about one week).

The lifetime of particles with the size below 20 nm is less than 20 hours.

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References
[1] Lushnikov A A, Zagainov V A and Lyubovtseva Y S 2015 Mechanisms of the formation of nanoaerosols in the troposphere Russian Journal of Physical Chemistry 34(10) 51–62
[2] Whitby K T 1978 The physical characteristic of sulfur aerosols Atmos. 12 135–59
[3] Baron P A and Willeke K 2001 Aerosol measurement: principles, techniques, and applications (New York) p 1172
[4] Air filters HEPA URL: https://vozdyx.ru/page/pylevye-filtry/ [Accessed 08.03.2021]
[5] Romanov N P and Zhukov G P 2000 Thermodynamic Relations for a Vapor Chamber Meteorol. and Hydrol. 10 37–52
[6] Rozenberg G 1983 Appearance and Development of Atmospheric Aerosol Bulletin of USSR Acad. Of Sciences. Phys. of Atm. and Ocean 19(1) 21–35
[7] Yermakov A N, Azoyan A E and Harutyunyan V O 2019 Air humidity effect on the formation of organic aerosol in the atmosphere J. Atm. and Ocean. Optics 32(2) 143–146