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CLOUDS AND AEROSOL IN STRATOSPHERE AND MESOSPHERE: POLARIZATION ANALYSIS AND TRENDS IN THE BEGINNING OF 21st CENTURY

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Abstract. The paper describes the polarization measurements of density and microphysical properties of stratospheric aerosol and noctilucent cloud particles and their relation with long-term trends of physical conditions of middle and upper atmosphere in a present time. It was shown that mean particle size of noctilucent cloud that directly defines its brightness and visibility increases in the case of mesopause drift to lower altitudes and expansion of the layer where water ice can be formed. This can explain the positive trend of cloud appearance rate in the case of zero temperature trend of mesopause. Stratospheric aerosol density shows the negative trend since 2011 to the summer of 2019, that is related with absence of medium and strong volcanic eruptions during this period. In late summer of 2019 upper troposphere and lower stratosphere were filled by aerosol possibly related with Raikoke volcano eruption on Kuril Islands in June 2019.

Mesospheric temperatures and noctilucent clouds

Increase of carbon dioxide in the atmosphere and greenhouse effect is now one of most actual problems of mankind. In the upper atmosphere where the density (total and of CO_2 in particular) is much lower, CO_2 can be optically thin in its infrared bands including the one at 15 μm, which is the basic for greenhouse effect and coincides with the range of maximal thermal emission of Earth. In this case the opposite effect can take place: shock excitation of CO_2 molecules in collision with other particles (mainly, with oxygen atoms) followed by their infrared emission leaving the atmosphere. This effect is called "radiative cooling" and can lead to temperature decrease of upper atmospheric layers [*Houghton*, 1970; *Roble and Dickinson*, 1989]. The process was actually detected [*Kokin and Lysenko*, 1994] and its value (that was overestimated as it was found later) reached –1*K*/year, about 50 times faster than global warming at the surface of the Earth!

More exact experimental and model analysis (for example, *Lübken et al.* [2013], Yuan et al. [2019]) gives the results depending of altitude and season, reaching about –0.2*K*/year in summer high-latitude mesosphere. It is known that fast upward air motion and adiabatic cooling makes this region coldest in atmosphere and entire Earth. Now the temperature can be as low as 130K or even less (see below). The reason of such trend is CO_2 which density increase there at 0.5% per year that is faster than in lower atmosphere [*Qian et al.*, 2017].

In a very beginning of 21st century the cooling of upper atmosphere had got slower. Models show that it can be related with stabilization of ozone content in stratosphere [*Akmaev et al.*, 2006; *Lübken et al.*, 2013]. In the same time the work of new missions on the atmospheric research had started. One of those is SABER (Sounding of the Atmosphere Using Broadband Emission Radiometry [*Russell et al.,* 1999]) instrument onboard TIMED (Thermosphere, Ionosphere, Mesosphere, Energetics and Dynamics, NASA) satellite. Techniques used there allow building the temperature and gas density profiles with very good vertical resolution (about 0.4 km). As we can see in Fig. 1 built basing on SABER June-July data for vicinity of Moscow, negative temperature trend remains in 2002-2019 in most part of layers not only in mesosphere but also in stratosphere.

Trend disappears only near the mesopause, at altitudes 82-87 km. This is interesting since ice crystals can form noctilucent clouds at these altitudes in summer. They were not observed in mid-latitudes until late 19th century, but now they are usually seen in June and July. Their visually estimated occurrence rate confidently increased during 20th century [*Thomas and Olivero*, 2001].

During the second half of 20th century, when satellite observations of mesosphere had begun, perennial mesospheric clouds were found around the poles, the field was expanding from year to year [*DeLand et al.*, 2017]. However, as we spoke above, there is no noticeable trend of temperature in the cloud layer (82 km). This is seen in Figs. 1 and 2, the latter shows the mean June-July temperatures in the vicinity of Moscow (±3° by latitude, ±15° by longitude) by TIMED/SABER [*Russell et al.*, 1999] and EOS Aura/MLS [Schwartz et al., 2008] satellite data together with results of twilight sky photometry and polarimetry [*Ugolnikov and Maslov*, 2013]. There is also no trend of H_2O by EOS Aura/MLS measurements at the same altitude. This contradiction and uncertainty of visual trends of cloud occurrence rate even led to conclusion that noctilucent clouds can not be used as display of mesosphere cooling [*Danilov*, 2012].

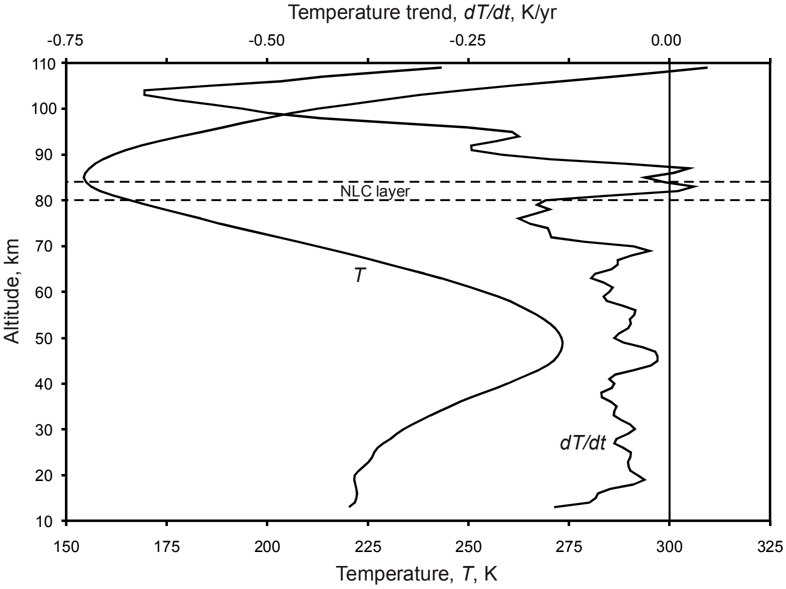

Figure 1. Profiles of mean June-July temperature and its trend in 2002-2019 in the vicinity of Moscow (±3° by latitude, ±15° by longitude) by TIMED/SABER data. The layer of noctilucent cloud (NLC) is shown.

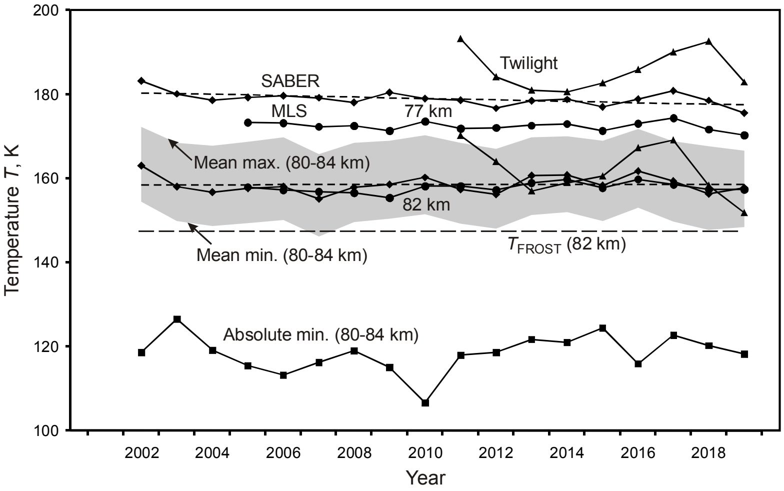

Figure 2. Mean June-July temperatures in 2002-2019 at 77 and 82 km in the vicinity of Moscow by SABER, MLS and twilight analysis. Absolute and mean minima and mean maximum in the layer 80-84 km by SABER data are also shown.

This problem can be solved if we measure not the cloud occurrence rate but physical characteristics, first of all, the mean particle size. This can help to find the principal factor for the appearance and brightness of clouds. For example, clouds in August 2015 and 2016 observed near the Polar Circle [*Ugolnikov et al*., 2016, 2017] were so bright but consisted of small or moderate particles (the mean radius is not more than 30 nm), pointing to the large particle number and probable relation with Perseids meteor stream.

In a present time, the particle size is measured by lidar sounding and from geophysical rockets and satellites (see references in [*Ugolnikov et al*., 2016]). It can be also found by polarization analysis of clouds if they cover the major part of the sky, i.e. wide range of scattering angles. It can be done by use of wide-field polarization sky cameras. These were also used for temperature profile retrieval in mesosphere. The technique of cloud field separation and finding the polarization is described by *Ugolnikov et al.* [2016], *Ugolnikov and Maslov* [2019a].

Bright noctilucent clouds were observed in central Russia at June, 25-27, 2018. Fig. 3 shows the dependencies of cloud polarization on the scattering angle compared with the one of clouds at July, 5, 2015 and Rayleigh dependency for very small particles (curve 1). We can see the significant difference between 2018 date and Rayleigh dependency. Observational data is much closer to model curve 2 corresponding to mean particle size 70 nm [*Ugolnikov and Maslov,* 2019a].

It is known that light scattering properties of small particles sharply depend on their mean radius (~r^6). Large particle size estimated by polarization analysis is well enough to explain the unusual brightness of clouds. This allowed rejection of hypothesis of large particle number and relation with Ozerki meteorite impact 300 km southwards from Moscow at June, 21, 2018.

To understand the possible reason of such particle growth, we have to look at thermal structure of mesosphere during those days. Here we should notice that widely used picture of mesosphere as a layer with monotonous temperature decrease to the minimum at mesopause is much simplified. Mesosphere is passed by the acoustical gravity waves originated in the lower atmosphere by non-uniform winds, cyclones and thunderstorms. These waves are seen as temperature variations in Fig. 4. Owing to this, temperature changes above and below the ice frost level that is also shown in this figure. This is the reason of visible wave-like structure of noctilucent clouds.

We see that temperature is often below the ice frost level, but clouds appear when these minima moves below "traditional" mesopause down to 80-82 km. This is related with larger density of water vapor. Shifting downwards through this cold and relatively humid layer, the ice particle can grow to radius 50 nm and more [*Rusch et al*., 2017]. Owing to sharp dependence of scattering by radius the latter value is principle for visible cloud formation. This is seen for temperature profile of bright cloud night of June, 25, 2018, when the frost layer was especially wide and its lower boundary shifted down to 78 km, lowest value for all profiles in Fig. 4.

This property removes the contradiction of trends of mesopause temperatures and noctilucent cloud occurrence rate. Although the summer temperature at 82 km is almost constant, it decreases below this layer (negative trends are seen for lower altitudes in Figs. 1 and 2) providing better conditions for large particle formation. This can be interpreted as shift of summer mesopause with constant temperature to lower layers with higher water vapor density. If this process continues, we may expect more often appearance of bright noctilucent clouds in summer night in future. However, we should remember that it is the reflection of increasing density of greenhouse gases in middle and upper atmosphere.

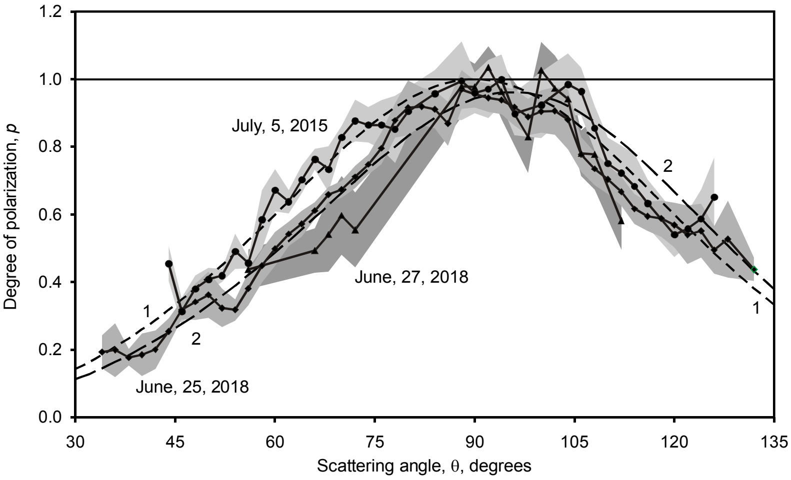

Figure 3. Polarization of light scattered by noctilucent clouds for 2015 and 2018 observations. Model *curve 1* corresponds to Rayleigh scattering (small particles), model *curve 2* is for lognormal particle distribution with mean radius 70 nm and distribution width σ=1.4.

Volcanic and background stratospheric aerosol

Warming process of lower atmosphere during last two centuries was sometimes interrupted by short cooling periods. "The year without a summer" in Europe and North America in 1816 is a good example. It became clear in 20th century that the reason of such cooling is volcanic activity, in particular, Tambora eruption in 1815 [*Humphreys*, 1920]. It was obvious that cooling was caused not by volcanic ash that fell on Earth quite fast, but by other volcanic-originated substance at higher altitudes expanding above the major part of the Earth.

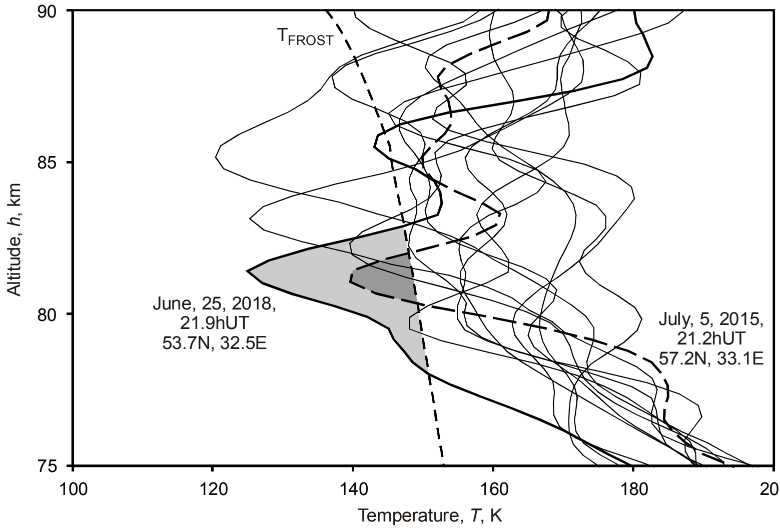

Figure 4. TIMED/SABER upper mesosphere temperature profiles in the vicinity of Moscow for 15-30 of June, 2018, compared with profile of NLC night in 2015. Ice frost temperature profile is calculated using empirical law by *Murphy and Koop* [2005].

First direct balloon measurements of stratospheric aerosol particles were conducted by *Junge et al.* [1961]. The chemical nature of the particles became clear 10 years later [*Rosen*, 1971], it is close to the clouds in the atmosphere of Venus. Particles are the droplets of sulfur acid. Reflecting the solar emission, they reduce it at the surface of Earth, decreasing the temperature. This is often called "anti-greenhouse effect".

Occasionally, first decades of intensive balloon study of stratospheric research [*Deshler et al.,* 2003] were the epoch of major volcanic eruptions: Agung, El Chichon and finally Pinatubo, the strongest eruption of 20th century in 1991. Effect of each eruption lasted for about 5-6 years. During the short intervals in between, concentration of aerosol did not vanish, remaining on constant background level [*Deshler et al*., 2006]. Detailed study of this background aerosol became possible since the very end of 20th century, with the relaxation of stratosphere after Pinatubo eruption.

During the first decade of 21st century, the optical depth of stratospheric aerosol had increased [*Solomon et al.,* 2011]. Possible anthropogenic reasons of this was discussed. It could be related with the increase of industrial release of SO_2, especially in South-Eastern Asia [*Lu et al*., 2010]. Another possible source of sulfur in stratosphere is carbonyl sulfide OCS [*Crutzen*, 1976]. Antarctic ice analysis had shown that its concentration is above its pre-industrial value [*Aydin et al.,* 2014].

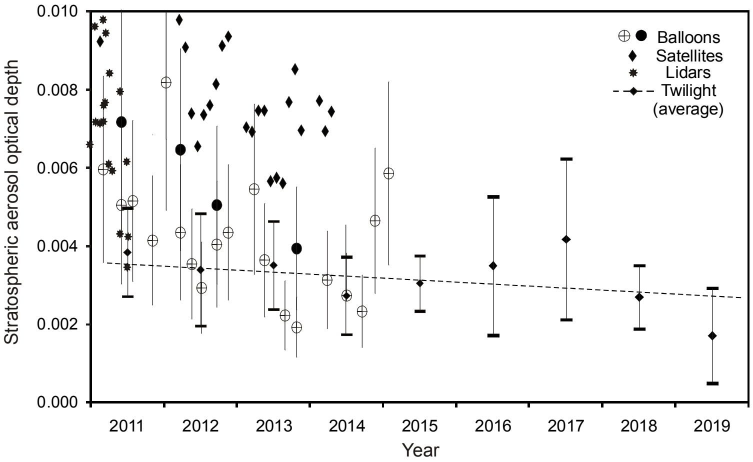

Figure 5. Yearly averaged vertical optical depth of stratospheric aerosol above 15 km for the wavelength 540 nm by twilight analysis compared with total vertical depth of stratospheric aerosol reviewed by *Kremser et al.* [2016].

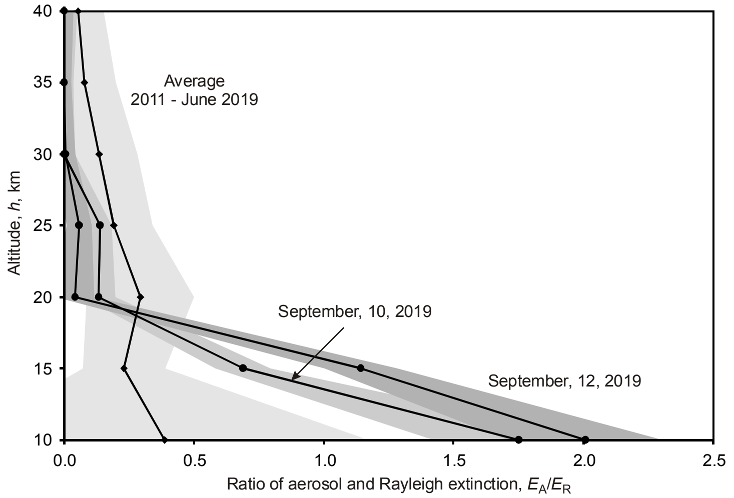

Figure 6. Profiles of ratio of aerosol and Rayleigh extinction in stratosphere averaged by volcanically quiet period until June 2019 and in the morning twilights of September, 2019.

However, there were some doubts on the anthropogenic nature of stratospheric aerosol increase in 2000s [*Neely et al.*, 2013]. The natural reason could be the moderate volcanic activity in a very beginning of 21st century, including the eruptions of Tavurvur, Kasatochi, Sarychev peak, Eyjafjallajökull, and Nabro. Following decade turned out to be more volcanically-quiet, and trend analysis of stratospheric aerosol can give the answer on this question.

Since the beginning of 2010s, different techniques of aerosol analysis (balloon, remote lidar and satellite sounding) had shown the decrease of optical depth of stratospheric aerosol [*Ridley et al.*, 2014, *Kremser et al.*, 2016]. Polarization all-sky measurements started in 2011 can be also used for the detection and analysis of stratospheric aerosol scattering, the method is described by *Ugolnikov and Maslov* [2019b]. Fig. 5 shows the yearly averaged values of stratospheric aerosol optical depth above 15 km at 540 nm compared with data reviewed by *Kremser et al.* [2016] for close wavelength. We see good agreement (with some offset of twilight data owing to boundary of integration above the tropopause, 15 km) and negative trend confirming the optimistic resume of *Neely et al.* [2013].

Data of 2019 in Fig. 5 is averaged on a first half of the year, until the end of June. Following was the effect of "purple light" during the twilight, that is typical for postvolcanic periods with increased aerosol level. Possible reason was Raikoke eruption at Kuril Islands in June 2019. Unfortunately, bad weather conditions in summer 2019 in central Russia restricted the value of observational data. However, we have obtained two aerosol profiles in morning twilights of September, 10 and 12. Fig. 6 shows the profiles of "aerosol to Rayleigh" extinction ratio for these twilights compared with average profile of 2011-2019 dates. Enhancement of aerosol extinction is significant, however, it is maximal in troposphere, at altitudes about 10 km, possibly expanding to lower stratosphere (note that Fig. 6 shows the ratio of aerosol and Rayleigh extinction, the Rayleigh value at 10 km is about twice of the one at 15 km). This result is visually confirmed by contrast shadows of vapor trail of plains on a purple twilight background observed in autumn 2019. So, the effect is mainly tropospheric, and additional check is needed to confirm its coupling with Raikoke volcano.

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