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ARTICLE

Evaporation and Fragmentation of the Electrified Droplets in the Polar Clouds during Spring Season as a Key Mechanism of the Ozone Depression Formation

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ABSTRACT

This study focuses on the role of the charged particles in the formation of the springtime ozone depression in the polar atmosphere. Analysis of experimental data collected in the polar atmosphere indicates that the small charged particles, predominantly ion clusters, can play a key role in the ozone molecules destruction and springtime ozone depression. The formation of these particles increases strongly during the spring season in the process of evaporation and fragmentation of the cloud-charged droplets in the lower stratosphere and upper troposphere. Additionally, small charged particles can also affect the formation and accumulation of chlorine monoxide under cold conditions of the lower stratosphere. At the same time, the chlorine mechanism of ozone destruction cannot completely explain the ozone depression formation, which probably takes place not only inside the polar vortex in the lower stratosphere but outside it also, both at the altitudes of the lower stratosphere and upper troposphere. The assumption that the charged particles play an important role in the process of ozone depression formation was put forward by previous studies, but in this research work, this assumption has been additionally confirmed, which is the springtime growth of ion clusters concentration as a result of the droplet's evaporation and defragmentation in the polar atmosphere is a key mechanism of the ozone depression formation. We simulated the process of evaporation and fragmentation in the case of a 10-micron size droplet, which implies the possible catalytic cycles of ozone destruction with the ion clusters. For the first time, the role of the Earth's magnetic field and the Polar vortex wind in the unipolar charge accumulation on the cloud particles in the lower stratosphere and upper troposphere not only inside the Polar vortex but also outside of it was substantiated. This fact in our point of view can give rise to the large-scale springtime ozone depression, spreading over the midlatitudes, which size is vastly greater than it is commonly supposed.

Keywords: Charged particles; Ozone depression; Polar Stratospheric Clouds (PSC); Condensation nuclei (CN); Evaporation and fragmentation of the charged particles; Ion clusters; Global Electric Circuit (GEC); Earth magnetic field

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1. Introduction

Usually, the accepted mechanism of the ozone depression formation in the polar stratosphere consists of ozone (O_3) destruction by halogens, and chlorine is the foremost of them [1]. The active chlorine which is necessary for ozone destruction is extracted mainly from so-called chlorine reservoirs HCl and $ClONO_2$ at their interaction on the surface of the Polar Stratospheric Clouds (PSC) particles [1,2]. Polar Stratospheric Clouds (PSC) are observed regularly during winter and springtime in the lower polar stratosphere, predominantly in the Antarctic [3]. The Arctic polar stratosphere is warmer in wintertime than that of the Antarctic, and that is why the Arctic PSCs exist during rather short periods of time as a rule [1–4]. Thus, the 'full-value' ozone holes are observed regularly in the Antarctic, and so-called mini holes are typical for the Arctic.

At the same time, in the previous studies [5–9], we showed that in the polar atmosphere, the small charged particles can play an important role in the ozone depression formation. It should be noted that the galactic cosmic rays are the source of the charged particles in the polar atmosphere. It is also important the ion formation maximal rate as a result of the galactic cosmic rays' arrival into the polar stratosphere is registered at an altitude of ~15 km [10] and very close to the ozone depression maximum. Also, a substantial contribution to the springtime ozone depression is made by the atmospheric dynamics thanks to which ozone from the tropical and mid-latitude regions is transferred to the polar area [3]. Sinking with cold air inside the polar vortex ozone reaches the altitudes of the lower stratosphere where the galactic cosmic rays affect the process of the charged particle formation and, as a consequence, the ozone destruction may be exposed to the most extent.

There are several observations about the important role of the charged particles in their effect on the ozone concentration in the Earth's atmosphere. One of the pioneering works where the ions and ozone interaction was studied by Morita and Ishikawa [11], which presented that there is an interaction between negative ions and ozone molecules that causes the vertical ion profile variations in the atmosphere. The relation between ozone concentration variations and cosmic ray intensity was detected rather long ago by many researchers who proposed various explanations for this fact. For example, Ruderman and Chamberlain [12] explained the decrease of the ozone concentration by the growth of nitric oxide content with the decrease of the solar activity and subsequently with the rise of the galactic cosmic rays intensity.

A new theory of the ozone hole formation was considered by Lu and Sache [13–15], who assumed that ozone holes may be a result of the free halogens released by the cosmic rays out of the polar stratospheric clouds (PSC) particles. Recently, Lu [16] gave evidence of the existence of the ozone depression in tropical regions that can change views on the concepts of the ozone hole formation and its danger level. According to Lu's opinion, the existence of such depression in the tropical zone can be explained only by the charged particles' participation in its formation.

The role of the charged particles in ozone destruction during solar flares is irrefutable. During solar bursts, the charged particles concentration rises in the atmosphere with the ozone concentration decreases [17,18]. For example, Solomon et al. [18] explained the effect of the ozone concentration drop in the mesosphere during the solar proton event in July 1982. In their point of view, the ozone concentration decrease is the result of the odd hydrogen formation during that event. The accumulation of the odd hydrogen brings the emergence of the so-called hydrogen cycle of ozone destruction.

According to our earlier analysis [5], the participation of small charged particles, mainly ion clusters, can give rise to ozone destruction in the polar atmosphere; chlorine oxide can accumulate, and many other chemical reactions can occur. However, from our point of view, these reactions go only on the surface of small charged particles and ion clusters but not on the surface of all PSC particles as believed in the classic theory of the ozone depression formation. The high efficiency of this type of reaction is confirmed by the theoretic estimations [5]. Also, these reactions have a set of specific features. Previously,

we estimated that in the process of collision with the small charged particles, not only a direct decomposition of various molecular species is possible, but also their chemical interaction with the nearly atomic and molecular surroundings of the ions and small charged particles. The principles of the formation of the atomic and molecular surroundings of small charged particles based on the features of the dipole interaction of charged particles with molecules are discussed in detail [5–7].

The most important compound in the atomic and molecular surroundings of the ions and small charged particles can be hydroxyl OH [5]. As a result of the independent interaction of the chlorine reservoirs HCl and $ClONO_2$ with hydroxyl OH in the surroundings of the charged particles chlorine monoxide ClO may be accumulated in space, and a set of chemical reactions can also take place [5]. At the same time, it is impossible in our opinion to explain the ozone depression formation only by chlorine mechanism.

As experimental analysis [5,7] showed, the effect of the small charged particles on ozone destruction may not be limited only by the area inside the vortex and lower stratospheric altitudes. The O_3 destruction can take place on the upper tropospheric altitudes as well as outside the polar vortex where chlorine compounds are virtually absent but small charged particles are present.

In the present work, we continued our previous research. The additional analysis together with the results which we obtained earlier allowed us to reveal the new features of springtime ozone depression formation. A more detailed analysis of the balloon measurements in the polar regions which were considered earlier, as well as experiments on evaporation and destruction of the charged particles allowed substantiate for the first time a hypothesis that it is evaporation and defragmentation of the charged particles in spring is a key mechanism of the ozone depression formation. In confirmation of this hypothesis, we carried out the simulation experiment of these processes on a case of a 10-micron size droplet.

The rate of the ozone molecules' destruction colliding with the small charged particles was estimated [5], but in the present work, the process of possible destruction of the ozone layer is verified. For the first time catalytic cycles of the destruction with the participation of the atomic and molecular surrounding of ions are considered.

As it was mentioned above, the main source of the charge which is accumulated on the aerosol and cloud particles in the polar atmosphere is the galactic cosmic rays. At the same time, the recombination of the charges of the opposite sign decreases the charge concentration [5]. Based on the experimental data and specific features of the altitudinal distribution of the charged particles we suggested and substantiated the assumption on the accumulation of the unipolar charges of different signs on the upper and lower surfaces of the cloud and aerosol layers due to the global electric circuit work [5,7].

In the present work, we considered for the first time the effect of the Earth's magnetic field on the charged particles moving inside the polar vortex. The vertical component of the magnetic field which is the strongest one in the polar regions also gives rise to the unipolar charge. In contrast with the electric field, which separates charges vertically, the magnetic fields separate them horizontally with the transportation of the negative charges outside the polar vortex and the positive ones inside it. In the outer part of the vortex where PSCs are absent, the charge is accumulated preferentially in the clouds and aerosol layers in the upper troposphere. Inside the vortex chlorine compounds can make a certain contribution to ozone destruction. We assume that these compounds are accumulated due to the interaction of chlorine-containing molecules with the small charged particles. Outside the vortex where chlorine compounds are practically absent, the ozone concentration variations are explained usually by its natural seasonal cycle.

For the first time, we substantiated, and showed the analysis of well-known experimental data, that the springtime ozone depression could be vastly greater in size than it is commonly supposed and can exist not only inside of the polar vortex, but also outside it in the middle latitudes over the territories

with the dense population. This fact may be to some degree hazardous to people because of the increased UV radiation background.

Note also a possible relation of the strong ozone depressions in the Northern hemisphere with the climatic change. Indeed, despite the emission decrease of the ozone-depleting substances since about 1994 as a result of the Montreal Protocol and its revisions execution record, ozone depressions are observed in the Northern Hemisphere. One may mention the ozone depressions of 2011 [19] and 2020 [20] which are comparable with the depressions in the Southern Hemisphere. In our opinion, that may be a result of the charged particle concentration growth.

According to our estimates [21–24], the fact is that during the last decades the charged particle concentration in the upper troposphere and lower stratosphere over the Northern territories of Russia and after 2000 even on the part of Russian territories in the midlatitudes increased significantly. This fact is associated with an unprecedented motion of the Northern magnetic pole from the Canadian Arctic ocean shore towards the northern Russian territories for about ~1000 km, and also with the peculiarities of focusing of galactic cosmic radiation by the Earth's magnetic field [21–24].

It ought to be remarked that the quantity of PSC during the last decades increased also over the Northern Russian territories and adjacent territories [25], and that can be associated with the increase in concentration of the charged particles over these territories [21–24]. Taking into account that the charged particles play a key role in ozone destruction, the ultraviolet radiation should increase to the Earth's surface due to the magnetic pole motion. These processes, along with other effects associated with the movement of the magnetic pole [26], maybe the cause of the natural climate change on the Earth.

The problem of climatic change due to the atmospheric vorticity variation and subsequently the charged particle concentration and ozone in the Earth's atmosphere which may affect the heating of the Arctic and atmospheric dynamics change needs further investigation. However, this problem steps outside the scope of this work. The results of this work need further detailed study and proof of our concept.

2. Materials and methods

2.1 Study areas

The research areas are the polar atmosphere over the Arctic and Antarctic and adjacent territories. The experimental data which was obtained from the balloons, satellite, aircraft, and ground-based experiments was used in this research work. The balloon experiments were conducted in Kiruna, Sweden (67.8°N, 20.2°E), and over the polar station McMurdo (77.8°S, 166.7°E). The Antarctic experiments were obtained on aircraft data. The aircraft ER-2 started its flight from the territory of Chile (53°S) toward Antarctica, reached the latitude of 78°S, turned around, and returned. The satellite data were obtained over the Arctic (2004/2005) and Antarctic (2005) inside the polar vortex [2]. Also, the averaged values of the parameters at the 50hPa level over the Arctic and Antarctic for the period 1979–2018 are based on the ERA-Interim reanalysis data [3]. The mid-latitude data were obtained from the measurements over the Russian territory: Tomsk (57°N, 85°E) and Obninsk (55°N, 36°E).

2.2 Data

The data for the balloon measurements are given in references [4,27,28,29]. The well-known available data obtained from the ER-2 aircraft flight on September 22, 1987, during the Airborne Antarctic Ozone Experiment (AAOE) were taken out of reference [30]. The spatial and temporal variation of the number of parameters in the Arctic and Antarctic, including the concentration change of chlorine oxide and ozone were considered on the basis of the satellite soundings (Aura MLS) in the Arctic for 2004/2005 and Antarctic 2005 [2], and the averaged data for the period 1979–2018 [3] were used in our research work. The measurement of the total ozone content in the midlatitudes over the Russian territory in 2011 [31]

was also used.

2.3 Research methods

The graphic matter and correlation analysis of the measurements of our interest were used for the validity test of our hypothesis. An important element of the research work was a cross-analysis of data, which was obtained from the various experiments. For example, we apply condensation nuclei (CN) as a tracer (indicator) of the presence of the small charged particles and keep track of their correlation with the ozone concentration at various altitudes, latitudes, and seasons using aircraft, balloons, and satellite experiments aiming at our hypothesis testing on a possible ozone molecules destruction in a process of their collision with the small charged particles. The CN concentrations vary in many experiments but the direct measurements of the ion and charged particle concentrations are very few. That is why CN is a handy tool for the analysis of the small charged particles' effect on the ozone concentration.

Then the conclusion that CN is a tracer (indicator) of the small charged particles' presence is made from the comparison of measurements of charged particles and condensation nuclei. (**Figure 1a**–**1f**). The analysis shows the altitudinal ranges of the increased concentrations of the small charged particle and CN coincide roughly, and the size ranges of the small particles and CN overlap. It is important also that these altitudinal ranges are on the upper and lower surfaces of the wide aerosol layer in the lower stratosphere and upper troposphere (**Figure 1e**–**1f**).

The comparison with other experiments shows that it is just in the springtime the evaporation and fragmentation of the charged particles begin, first of all on the upper and lower boundaries of clouds and aerosol layers, which give rise to the significant growth of the ion clusters. The CN, formed in spring, are the ion clusters. Thus the applied cross-analysis brings a new understanding of the processes in the Earth's atmosphere. A number of theoretic calculations were carried out also. We simulated a process of the evaporation and fragmentation of the 10-mkm size droplet which allowed to substantiate a possibility of a significant increase of the ion clusters concentration in the polar atmosphere in springtime. Along with that, the theoretic estimations of the magnetic field effect on the moving charged particles inside the polar vortex allowed us to relate the density distribution of charged particles inside the polar vortex and outside it with the specific features of the wind velocity gradient distribution. On the basis of the estimations of the mutual effect of the electric and magnetic fields, some specific features of the charged particle distribution on the upper and lower cloud boundaries in the upper troposphere and lower stratosphere are revealed. Possible catalytic cycles of the ozone destruction with the participation of the charged particles were proposed and the destruction rate of ozone molecules in the process of their collision with charged particles was estimated.

3. Results and discussion

3.1 The relationships between condensation nuclei, charged particles and ozone destruction in the polar atmosphere

Specific properties of the unipolar charge accumulation on the PSC particles as a result of the global electric circuit operation

The global electric circuit operation is described by Harrison et al [32]. In cloudless weather weak currents ~ $2pA/m^2$ flow down from the lower ionosphere (~80 km), which is charged positively relative to the Earth's surface [32]. With the emergence of cloudiness which acts as a kind of resistance to electric current the unipolar charges are accumulated on the upper and lower cloud surfaces, positive and negative ones correspondingly. The cause of the unipolar charge accumulation is a variation of charged particles' mobility inside and outside the cloud [5]. Relatively small particles and ion clusters with a relatively high mobility are involved predominantly in the current system outside the cloud. At the same time, the charged particles grow in size inside the cloud due to the liquid condensation on them and also because of the process of adhesion of small particles and ions to aerosols and cloud droplets, and that is why their

mobility decreases [5].

However, the effects of accumulation of charged particles at cloud boundaries discussed above are considered in the research literature concerning the lower tropospheric clouds [32]. As our analysis performed earlier [5,7,8] showed, the global electric circuit operation is significant not only in the lower troposphere but also in the upper troposphere and stratosphere as well. The main source of the charged particles is the galactic cosmic rays. The estimations of the accumulated charge in the upper troposphere and lower stratosphere based on the balloon experiments are given in reference [5]. The concentration values of the accumulated charge on the horizontal cloud borders can reach ~10^3–10^4 *e*/cm^3 with the depth of the charged layer of ~1 km [5].

The detailed results of the balloon experiments in the polar Arctic atmosphere are illustrated in **Figures 1a–1d**, showcasing the measurements of charged particles in Kiruna, Sweden (68ºN) as outlined in reference [27]. Renard et al. [27] utilized an aerosol counter with special traps to eliminate charged particles from the airflow for concentration measurements. **Figure 1a** displays the vertical profiles of aerosol concentrations with and without the traps during balloon motion, revealing the profile of charged particles by subtracting the two. **Figures 1b–1d** depict these profiles for different charged particle sizes, indicating a concentration of charged particles in the upper and lower parts of the wide aerosol layer at altitudes of ~6–25 km. Our analysis suggests that positive charge is concentrated in the upper cloud part, while negative charge is concentrated in the lower part [5].

Balloon Measurements Showed: Ozone Destruction Takes Place in The Regions with Increased Concentration of Condensation Nuclei

In the vicinity of upper and lower cloud and aerosol layers borders not only relatively large charged particles were detected but also increased concentration of the condensation nuclei (CN) (**Figure 1e** and **1f**) [4]. Taking into consideration that CN size range is 0.02–1 μm [28,33] and it overlaps partly with the size range of the smallest charged particles (0.35–1 μm), which were determined in the experiment of Renard et al. [27] It is logically to assume that CN is also charged. This weighty conclusion is confirmed by other observations which will be considered further on.

The condensation nuclei are observed regularly in winter and springtime in the presence of clouds and aerosol layers [28]. The results of the balloon measurements of aerosol, CN, and ozone concentrations on January, 23 and 30, 1989 over Kiruna are shown in **Figure 1e–1h** [4,28]. The results of the balloon measurements of CN in various seasons in Antarctic are shown in **Figure 1f**.

As can be seen in **Figure 1a–1h** an increased CN concentration is observed at the upper and lower borders of the wide aerosol layer in the altitude range of ~6–25 km, where the reduced ozone content is also detected. Note, that ozone concentration decrease is observed both at the upper aerosol layer border (**Figure 1g**), and the lower one, as can be inferred from the plotted vertical profile of the ozone partial pressure in **Figure 1h**. It will be obvious from the further representation that the effect of the ozone concentration decrease while the CN concentration increases is caused, at least partly by the direct ozone destruction in the process of collision with small charged particles, and CN among them.

As was mentioned above, the CN size range which was measured by aerosol counters, which were installed on balloons and the ER-2 aircraft (see below) is of 0.02–1μm [28,33]. At the same time the charged particles' potential increases with their size reduction, and a significant ozone volume, according to our hypothesis, can be destructed by particles of smaller size, i.e. ion clusters of nanometer range [5]. Their concentration in the lower stratosphere can reach the value ~$(1–5)\cdot10^3$ cm^{-3} [10,11,34], which exceeds drastically CN concentration measured by this counter. However, it is important that CN act as tracers (indicators) of the presence of the small charged particles, including ion clusters [5–9].

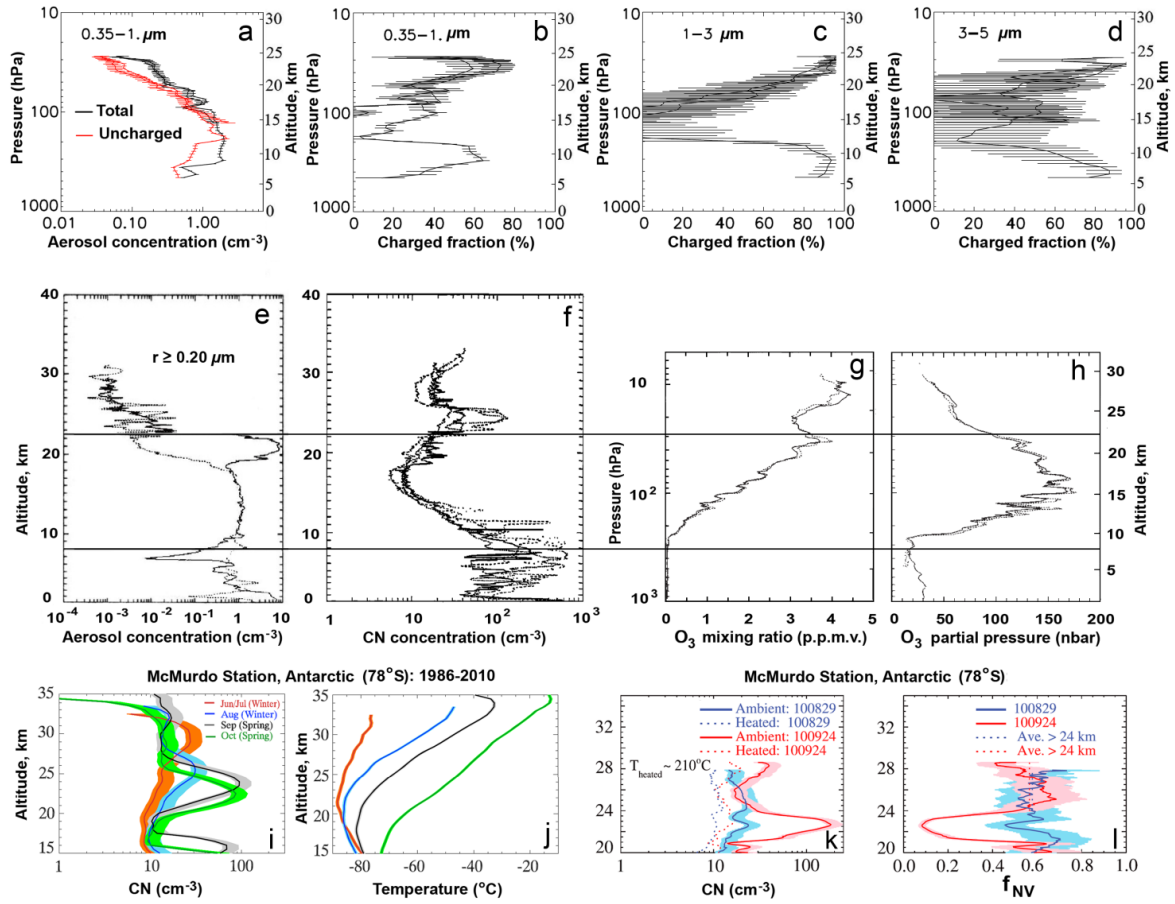

Figure 1. The balloon measurements: (**a**) Vertical profiles of the total aerosol concentration and uncharged aerosol concentration in the 0.35–1 μm range during upward and downward balloon motion over Kiruna on March 12, 2011; (**b–d**) percentage of the charged aerosol for various charged particles sizes; (**e**) vertical aerosol profiles over Kiruna on January 23, 1989 (solid line) and January 30, 1989 (dashed line); (**f**) vertical profiles of the CN concentration over Kiruna, January, 30, 1989 (solid line) and in Antarctic, 78ºS on August, 27, 1988 (. . . .); September 6,1988 (…..); September 14, 1988 (----); (**g–h**) vertical profiles of the ozone mixing ratio and partial pressure, correspondingly, over Kiruna on January 23, 1989; (**i–j**) mean values of CN concentration and temperature, correspondingly for winter months and spring months averaged for the period 1986-2010 over McMurdo Station; (**k**) CN volatility measurements on McMurdo station: the vertical profile of CN variation during their heating in winter on August 10, 2010 (blue lines) and in spring on September 9, 2010 (red lines); (**l**) profiles of non-volatile fraction of CN particles for the same dates as for (**k**).

Sources: Hofmann et al [4, 27–29].

Considering that the destruction of ozone molecules can occur with the participation of small charged particles with a significant surface electric potential [5–7], the processes of adhesion to larger particles and condensation on small charged particles, including ions, actually block the destruction of ozone inside the cloud. **Figure 1e, 1f** and **1h** show that maximal ozone concentrations are detected at the approximately minimal CN concentration in the middle of aerosol layer.

Seasonal variations of condensation nuclei represent the dynamics of ozone depression formation

According to our analysis given above and considered in detail in reference [5], the destruction of O_3 and a set of chemical reactions, including reactions of the free chlorine release from the chlorine reservoirs can take place at the upper and lower borders of aerosol layers and PSC in a relatively narrow altitude ranges as a result of the Global Electric

Circuit operation. These ranges correspond to the increased CN content. As it was mentioned earlier, CN is the tracer of the small charged particles which participate, in our opinion, in ozone destruction. The upper-level range where CN in winter is detected corresponds to the altitudes of 22–28 km, and the lower range which is more 'blurred' lies on the upper tropospheric altitudes, about ~3–10 km (**Figure 1f**). A possible cause of such 'blurring' of the lower CN altitude profile is a 'jaggedness' of the aerosol profile at these altitudes taking into account that inside the aerosol layers the adhesions of CN to aerosol particles grow stronger.

An important result of the balloon observations in the Arctic and Antarctic obtained by Hofmann et al. was the detection of CN concentration growth in springtime [4,28]. Their experiments showed that this growth began under conditions of direct solar illumination. Hofmann et al. [4,28] put forward a proposal that the formation of CN and their concentration increase in spring above ~18 km is related to the activation of the photochemical processes and homogenous or ionic nucleation of HNO_3 and water vapors [28]. Besides this, it was shown in reference[28] that there is a difference in the altitude position of layers of the increased CN concentration depending on the time of observation at the site of McMurdo Antarctic station (78º S).

Further investigations of CN dynamics showed [29] that along with an increase in CN concentration in the spring season, there are significant changes in the altitude position of the CN layers in the polar atmosphere. **Figure 1i** shows the average vertical profiles for the period 1986–2010 of the CN concentration for the winter months (June–July, August) and for the spring months (September, October) at McMurdo station [29].

It should be noted that the layers of the increased CN concentration in the polar atmosphere were detected not only inside the polar vortex but also outside it [33]. The CN concentration grows rapidly outside the polar vortex with altitude decrease (below 20 km); its maximum is observed at altitudes ~8–10 km [33]. This result agrees with the observations of Hofmann et al. [28] of the lower CN layer (**Figure 1f**), as well as with the results of the SOLVE experiment [5,35]. In the March 11, 2000 experiment during the ER-2 aircraft 'dip' from 20 to 15 km outside the polar vortex a strong CN concentration rise [5] was observed which was accompanied by a sharp O_3 drop. A similar effect but during the 'dip' inside the polar vortex is also observed in the airborne experiments in the Antarctic [5].

The detailed results in Figure 1i shows that alongside with the strong increase of the CN concentration from winter to spring, there is a rather rapid change in the position of the increased CN concentration layers, particularly of the upper layer. The analysis of temperature variations in winter and springtime at McMurdo (**Figure 1j**) allowed us to put forward a proposal that a rapid sinking of the upper CN layer is associated mainly not with the sinking of clouds but with their vanishing, or evaporation, first of all of the upper cloudiness where temperature growth in spring is the most significant. This assumption explains some peculiar processes in the polar atmosphere.

The last remarks in fact clear up the essence of condensation nuclei. Hofmann's [4,28] assumption that the formation of CN and the increase of their concentration in spring is associated with the activation of photochemical processes and homogenous or ionic nucleation of HNO_3 and water vapours is only partially correct. The CN formation according to our analysis is associated with ionic nucleation, but not with the homogenous one. In fact, condensation nuclei which were discovered by Hofmann et al. [28] are rather large ion clusters, moreover, both water and acid molecules like HNO_3 and H_2SO_4 are present in the surrounding ions. Also, in our opinion, the springtime CN concentration rise is due to ions emission in the process of evaporation and fragmentation of the larger charged droplets [36–41].

Considering that the accumulation of charges in aerosol and cloud layers in the polar stratosphere occurs, first of all, at the horizontal boundaries of these layers as a result of the GEC operation, there is not just an evaporation of droplets in these areas in springtime, but the evaporation of charged droplets during their fragmentation, accompanied by ion

emission in the certain conditions [36–41].

A partial confirmation of the fact that the springtime CN are the ion clusters is the investigation of their volatility. One may see that heating up to ~ 210°C of CN formed in spring at altitudes ~ 21–24 km brings to their almost complete vanishing. (**Figure 1k**), where the relative concentration of the nonvolatile fraction decreases by almost an order of magnitude (**Figure 1l**).

In fact, the dynamics of CN concentration variation given in **Figure 1i** corresponds to the process of ozone depression formation in the lower stratosphere and upper troposphere. The gradual PSC disappearance is due first of all to the temperature rise (**Figure 1j**), accompanied by the filling of the greater part of space where PSC and upper tropospheric clouds existed earlier by the small charged particles and ion clusters. With the participation of these small charged particles, the destruction of ozone and the formation of ozone depression occur.

The analysis of data presented in **Figure 1** shows that in a process of the ozone depression formation, it is possible to separate rather conditionally tropospheric and stratospheric contributions which correspond to the lower and upper borders of a wide aerosol or cloud layers in the polar atmosphere predominantly in winter and early spring (**Figure 1a–1h**). As our further analysis shows the stratospheric contribution may be absent in the absence of PSC, but at the same time the tropospheric contribution may be quite notable both inside and outside the polar vortex in the presence of cloudiness in the upper troposphere. In this case, the accumulation of charge on cloud particles in the upper troposphere occurs in the same way as in the lower stratosphere, with the participation of the GEC and possibly the Earth's magnetic field.

Definitely, our assumptions need further verification and testing.

3.2 Peculiarities of Evaporation and Fragmentation of Charged Particles

Let us consider in more detail the processes of evaporation and fragmentation of charged droplets, taking into account the importance of these processes in the formation of the ozone depression in the polar atmosphere.

The process of the droplet evaporation was studied in various papers [36–41]. When a droplet evaporates and reaches the Rayleigh limit it begins to fragmentize into daughter particles, the number of which can range from 1 to 9 according to Roth et al [40]. The evaporation of the daughter particles and their reaching to the Rayleigh limit causes further fragmentation of the daughter particles. Consider the process of fragmentation of the charged particle with a radius of 10μm and a charge of $3.2x10^{-17}$ C. That is the average charge of the droplet with a given size in clouds. The relation between the particle size and its charge is given in **Figure 2a**. Meanwhile, **Figure 2a** also shows shows the maximal possible charges of droplets and solid particles as well as an average droplet charge for various particle sizes [5].

The size decrease of the evaporated particle is marked by the black line. The crossing of this line by the blue line means the reach of the Rayleigh limit when the droplet begins to fragment. We consider in our calculations the fragmentation of a droplet into two daughter droplets with equal charges. In the process of the further reaching of the Rayleigh limit, and this corresponds to one step in a black line change, more and more charge droplets appear, and their total potential exceeds substantially the potential of the initial droplet. This process finishes with the formation of a droplet of a nanometer range which contains one elementary charge.

However, in the processes under consideration, there are peculiarities which have relation to the formation of the ion clusters at a certain stage of evaporation, when a droplet reaches the nanometer size range. These peculiarities were studied in references [36–39,41]. The conclusions, which were based both on experiments and computer simulations [36–39,41] show that when a droplet achieves the nanometer size range and reaches the Rayleigh limit, the emission of ion clusters occurs. According to Consta et al. [37], these clusters include about ~10 molecules of water (**Figure 2b**).

In **Figure 2c**, we present our calculation of a scenario of ion emission due to the evaporation and

fragmentation of a 10 μm radius size droplet with a charge of 3.2×10^{-17} C. One may see in **Figure 2c** that the evaporation and fragmentation of this droplet results in the formation of about 200 ion clusters.

The ion clusters and small charged particles which are released from the evaporating cloud particles can accumulate on the clouds and aerosol layers boundary borders as a result of the GEC operation. As mentioned above, the concentration of the accumulated charge on the horizontal cloud boundaries for a 1 km layer can reach values of ~10^3–10^4 *e*/cm^3 [5]. As estimations given below show, such a charge can cause a significant ozone depression. Note that the concentration of charged particles can also be influenced by the Earth's magnetic field acting on charged particles moving in the polar vortex.

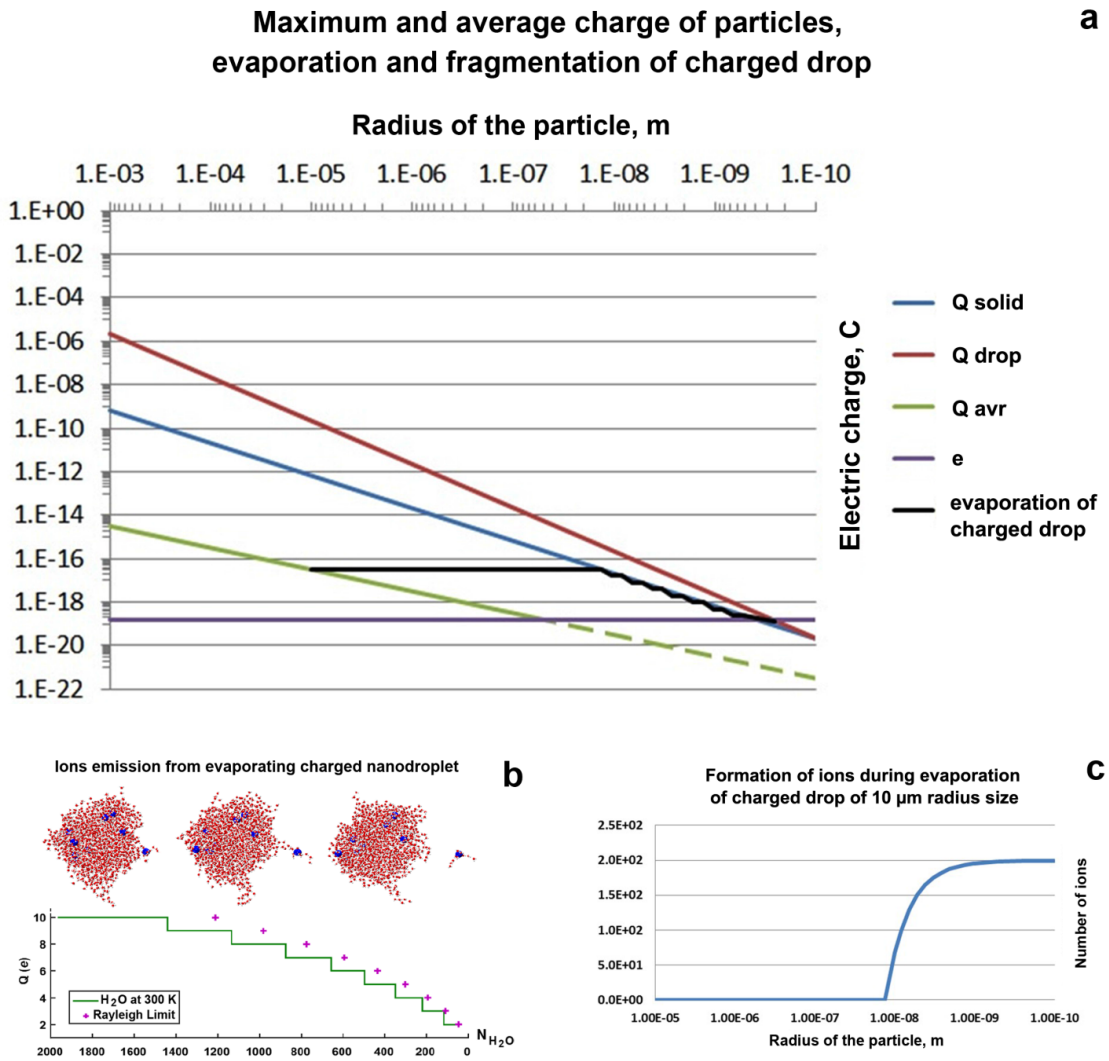

Figure 2. (**a**) A ratio between ionic charge and particle size for the strongly charged solid particles (blue line) and maximally charged water droplets (red line); and the average charge for tropospheric cloud particle (green line). The relationship between ionic charge and particle size is extrapolated for sizes less than 1×10^{-9} m. A straight purple line corresponds to the elementary charge of 1e. A process of the evaporation and fragmentation of a droplet with a charge 3.2×10^{-17} C (average charge on a particle of a 10 μm radius size) is shown (black line). Upon reaching a Rayleigh limit a particle fragmentizes into two parts with equal charges. (**b**) The process of evaporation of a droplet with a nanometer size range (about 2000 water molecules and a charge of 10e. Upon reaching a Rayleigh limit, the ion cluster of about ~10 water molecules is ejected; (***c***) The formation of ion clusters in a process of evaporation of 10 μm radius particle with a charge of 3.2×10^{-17} C when it reaches Rayleigh's limit.

Sources: Belikov et al [5, 37].

3.3 Catalytic cycles and ozone destruction rate with participation of small charged particles

Ozone molecules as well as many other chemicals can be destructed on the effective surface of ion cluster [5]. The dissociation of these chemicals including ozone can initiate a set of chemical reactions in the surroundings of the charged particle. The products of these reactions with various dipole moments can either erupt into the environment or stay in the near ion surrounding. The principles of these reactions are given in detail in my previous study in 2016 [5].

According to our concept, a majority of the heterogeneous reactions in the polar atmosphere proceed only on the effective surface of the small charged particles, mainly ion clusters, and not on the surface of all PSC particles. The charge on the particles accelerates this reaction, making their existence possible.

The chain of chemical reactions involving small charged particles begins with the appearance of hydroxyl in the environment of these particles (equations (1)–(4)):

$$O_3(v) \rightarrow O_3(s) \quad (1)$$

$$O_3(s) + M(s) \rightarrow O_2(s) + O(s) + M(s) + 1.4\ eV \quad (2)$$

$$O_2(s) \rightarrow O_2(v) \quad (3)$$

$$O(s) + H_2O(s) \leftrightarrow 2OH(s) \quad (4)$$

Where s and v denote the locations of molecules: ambient space is v (volume), and the surface of a charged particle is denoted by s (surface).

According to estimates given [5], an O_3 molecule in a process of collision with ions of H_3O^+ type can exceed a threshold of ~1 eV and decompose into an oxygen molecule and hydrogen atom on the ion effective surface (1)–(2). Then an O_2 molecule which has a zero dipole moment will erupt into the environment with a high probability because of the absence of the attraction to the ion (3). At the same time, an O atom can enter into a reaction with a water molecule though the atomic oxygen also has a zero dipole moment (4).

Analysis shows that the O atom has exceptional chemical activity on the effective surface of charged particles, one of the reasons for which is the possibility of using the energy of ~1.4 eV released in the ozone destruction reaction (2). It is possible that rapid bi-lateral reactions can support the existence on the charged particle surface both hydroxyl molecules and oxygen atoms. A permanent presence of either the O atoms or hydroxyl OH molecules in the surroundings of the charged particles provides a possibility of catalytic oxygen and hydrogen cycles and also a mixed oxygen and hydrogen cycles of the ozone destruction.

It should be noted that molecules of HOONO and $HOONO_2$ can also be a source of hydroxyl in a neighbourhood of ions and small charged particles.

Let us consider catalytic cycles of ozone destruction for both positively and negatively charged ions which contain water molecules.

The hydrogen cycle suggests the presence of the hydroxyl molecules in the surroundings of ion. The hydroxyl molecule OH originates from the reaction (4). Here is the cycle with a participation of the ion H_3O^+ (see equations (5) and (6)):

$$O_3 + (H_3O^+)(H_2O)_n(OH) = O_2 + (H_3O^+)(H_2O)_n(HO_2) \quad (5)$$

$$O_3 + (H_3O^+)(H_2O)_n(HO_2) = 2O_2 + (H_3O^+)(H_2O)_n(OH) \quad (6)$$

Then the reactions recycle, and destruct more and more ozone molecules.The catalytic cycles of the ozone destruction can proceed with the participation of both positive ion clusters and negative ones.The oxygen catalytic cycle with the participation of the negatively charged O_2^- and O atoms availability in the ion surroundings are in equations (7) and (8):

$$O_3 + O_2^-(H_2O)_n = O_2 + O_2^-(H_2O)_nO \quad (7)$$

$$O_3 + O_2^-(H_2O)_nO = 2O_2 + O_2^-(H_2O)_n \quad (8)$$

Evidently, that also a mixed oxygen and hydrogen cycle can exist, when ozone molecules can collide ei-

ther with the hydroxyl molecule, or with oxygen atoms.

Note that lifetime of molecules on the ion effective surface is extremely short. Thus, it is estimated that the ozone lifetime in the reaction (5) is about ~ 10^{-8} s, or less. Therefore, the lifetime of ozone in space, provided that it is destroyed on the surface through the considered catalytic cycles, is determined by the time of "pumping" of ozone to the effective surface of charged particles. This time for ozone was estimated by us [5]. The lifetime of ozone and some other compounds is to be determined from the following equation (9) [5,42]:

$$t = (PSvn / 4)^{-1}, \quad (9)$$

where P is a probability of the 'adhesion' of a molecule to the ion; S is an effective square of ion, n and v are ion concentration and average velocity of the compound molecules, consequently.

Assuming that ion concentration varies in the stratosphere in the range $(1–5)\cdot10^3$ ions/cm^3 [10,11,34] and the effective radius of 'adhesion' is ~10Å (for H_3O^+) [5], one can estimate ozone lifetime. For the polar stratospheric conditions it is from 10 days to 2 months if P=1 and given ion concentration variation. In this case a rate of the ozone destruction *k* lies in the range $(2.4\cdot10^{-7}–1.2\cdot10^{-6})$ 1/s, where *k* = PSvn/4.

The ozone depression formation in the polar stratosphere is registered in the spring months, that is the ozone lifetime is of the order of one month. Taking into account our estimates, we concluded that the small charged particles could provide very likely the ozone depression formation. It means that the effectiveness of the proposed mechanism of ozone destruction is highly competitive with that of the conventional chlorine mechanism which is considered as responsible for the ozone depression formation.

3.4 Effect of the magnetic field and Polar vortex wind on the distribution of charged condensation nuclei and ozone

When a charge moves in the electric and magnetic fields the Lorentz force F arises in equation (10):

$$F= q(vxB + E) \quad (10)$$

where q is the electric charge of a particle, v is a particle velocity, and B and E are magnetic and electric field strength, correspondingly. As the strength of the vertical electric field component at the Earth's surface is ~100 V/m, and a contribution of the magnetic field component for typical wind velocities at the Earth's surface is only ~0.01 V/m, the latter parameter effect on the charged particles is usually neglected for the lower troposphere.

However, with the altitude rise the strength of the electric field decreases and wind velocity and a share of the magnetic component contribution to the Lorentz force rises.

Calculations show that in the polar stratosphere the vxB product can reach ~0.1 V/m for a wind velocity of ~230 km/h. This velocity is quite typical for stream flows including the Polar vortex. Under these conditions the vertical component of the electric field is about ~0.5 V/m for the altitude of 20 km, i.e. it is comparable with the magnetic component of the Lorenz force.

Thus, under the Polar vortex conditions in the lower polar stratosphere the magnetic field effect on the distribution of the charged particles should not be neglected.

Let us consider now how the magnetic field in the Polar vortex condition acts on the charged particles generated by the galactic cosmic rays. For our first developed qualitative analysis we shall neglect the horizontal component of the magnetic field taking into consideration that the vertical component of the magnetic field exceeds greatly the horizontal one.

The results show that under these conditions in the upper troposphere and lower stratosphere the positive charges under the magnetic field influence are focused inside the Polar vortex and negative charges are transported outside it. **Figures 3a** and **3b** show basic schemes of this distribution in Arctic and Antarctic.

In **Figure 3c** and **3d** a principle scheme of interaction and effect of the electric and magnetic field on the charged particle motion in the Polar vortex condition is shown. Two cases are considered. The first case, is when the cloudiness and aerosol layer are present in the polar upper troposphere and stratosphere, and the second case: the clear atmospheric conditions.

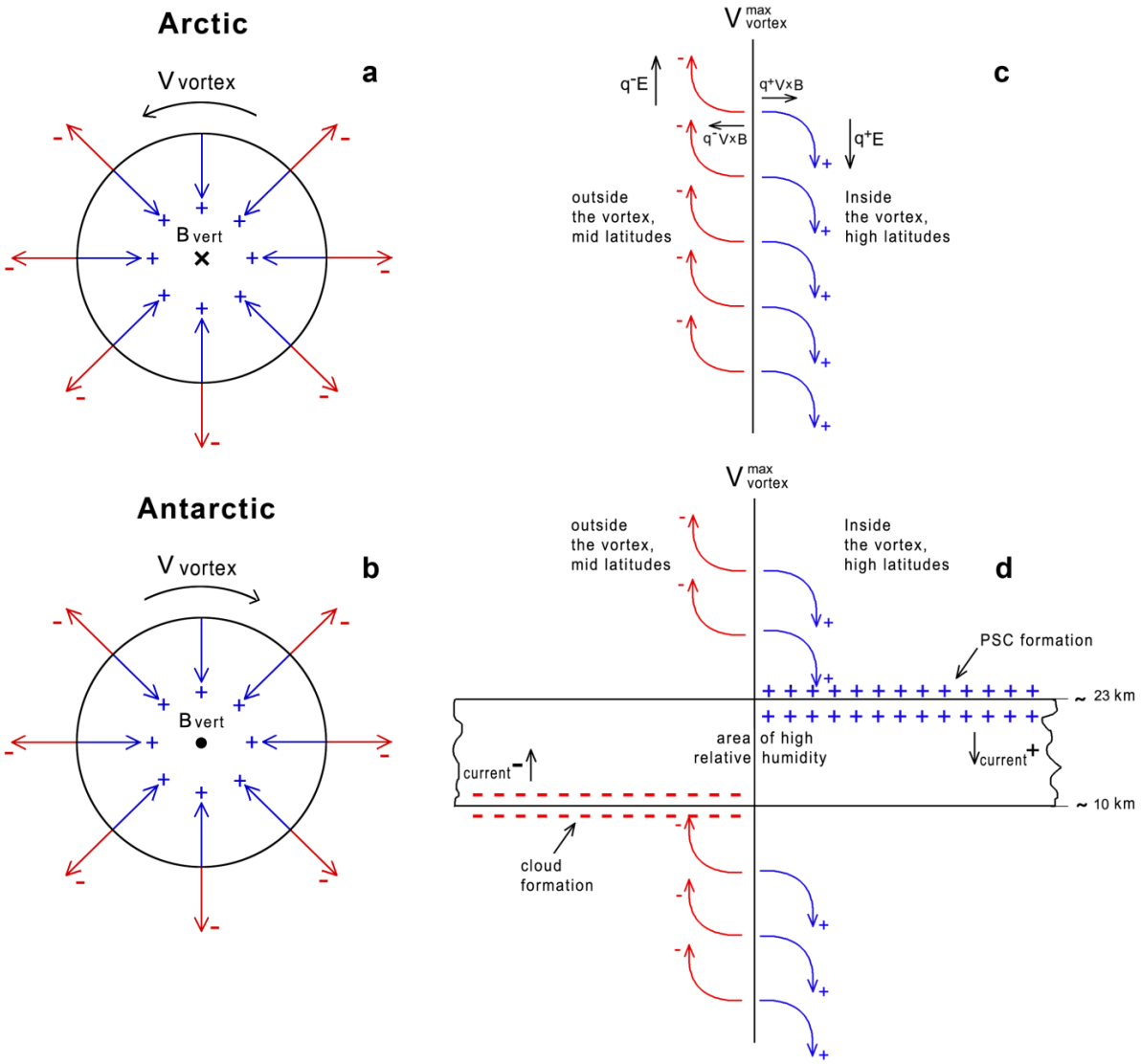

Figure 3. A principle scheme of the electric and magnetic fields effect on the charged particles motion under the conditions of the Polar vortex. The positive charges are focused into the inner side of the polar vortex under the effect of the vertical component of the magnetic field and wind; the negative charges are transported into the outer part of the vortex: (**a**) Arctic; (**b**) Antarctic. (**c**) and (**d**) are cases for a pure atmosphere and atmosphere with clouds or aerosol layer in the upper troposphere and lower stratosphere, correspondingly. In case (**c**) the unipolar charges are trapped by the electric field and give rise to electric currents in various directions both inside and outside the vortex. In case (**d**) the charges are accumulated inside the clouds and create abundance of the positive and negative charges on the given layers, which contributes to cloud formation in these areas.

In the case of the absence of cloudiness or aerosol layers the negatively charged particles which are transported outside the vortex as a result of the influence of wind and magnetic field on them, are caught up by the vertical electric field and creating the additional negative upward current. Similarly, the positively charged particles give rise to the additional positive downward current (**Figure 3**c).

In the presence of the cloud or aerosol layers ions and small particles adhere to the larger particles of these layers or become larger particles as a result of liquid condensation on them. With that layers of the unipolar charge emerge in the upper part of clouds inside the vortex (positive charge) and lower part of them outside the vortex (negative charge) (**Figure 3d**).

It should be noted that as a result of the GEC operation a unipolar charge is accumulated also the horizontal cloud borders: the positive charge on

the upper border and the negative one on the lower border [32]. Evidently, a sign and excess of the total charge which is accumulated by magnetic and electric fields effect with taking also the process of recombination will correspond to the distribution shown in **Figure 3d**.

It is necessary to mention also possible feedback between specific features of the charge particle distribution and cloud formation in the polar atmosphere.

The unipolar charge concentration increase inside and outside the Polar vortex can promote the cloudiness and aerosol layers formation in these areas in a ~ 10–20 km altitude range in accordance with the peculiarities of the vertical profiles of the temperature and humidity. However, while the Polar vortex exists the processes of the formation and further sustain of the upper part of clouds and aerosol layers inside the vortex and lower part of them outside the vortex are strengthened (**Figure 3d**). According to estimates in our previous studies [5], the concentration of the accumulated charge at the cloud borders by just only the GEC operation may vary in a wide range: ~ $2 \times 10^2 - 2x10^5$ e/cm^3.

Let us consider now how the proposed model of the magnetic field and Polar vortex wind agree with the experimental data which were obtained in the aircraft experiment on September 22, 1987 under the program AAOE 87 [30] (**Figure 4**).

ER-2 aircraft in this experiment flew at the ~20 km altitude. This altitude corresponds approximately to the upper border of the PSC [5], where, according to our estimates, a layer of the mostly positively charged particles should be concentrated (**Figures 1a –1d**). Subsequently, these particles under the influence of the magnetic field and vortex wind should be redistributed according to our model (**Figure 3b**).

The Lorentz force (10) which affects the charged particle can be decomposed into horizontal and vertical components and can be rewritten as equation (11):

$$F = q\,(vxB_{vert} + E_{hor} + E_{vert}) \tag{11}$$

Here, B_{vert}, E_{hor} and E_{vert} are the corresponding vertical and horizontal components of the magnetic and electric fields. In equation (11), as it was mentioned above, we neglected the horizontal component of the magnetic field.

Let us assume that there is a quasi-static equilibrium of charges along the aircraft's horizontal flight path. This condition can be written as equation (12):

$$dF/ds = qB_{vert}(dv_{hor}/ds) + dE_{hor}/ds + dE_{vert}/ds = 0 \tag{12}$$

Assuming that in the horizontal direction as equation (13):

$$dE_{vert}/ds = 0 \tag{13}$$

we shall obtain equation (14):

$$qB_{vert}(dv_{hor}/ds) + dE_{hor}/ds = 0 \tag{14}$$

Basing on equation (14), we can obtain the following result in equation (15):

$$qB_{vert}(dv_{hor}/ds) = -dE_{hor}/ds = \rho/\varepsilon_0\ , \tag{15}$$

where ε_0 is the dielectric constant.

The right part of the equation (15) is in fact a Poisson equation for the one-dimensional problem as equation (16):

$$dE_{hor}/ds = -\rho/\varepsilon_0 \tag{16}$$

Referring to the equations (15) and (16) we obtain an important conclusion as equation (17):

$$ABS(dv_{hor}/ds) \sim \rho \tag{17}$$

Figure 4 shows the map and altitudes of the aircraft flight as well as results of the data analysis for September 22, 1987 during AAOE expedition [30], which include, based on publicly available data [30], distributions along the aircraft path of a number of physical values.

Figure 4d shows a calculated absolute value of the average wind gradient along the aircraft flight path. The period of the gradient averaging was 500 sec. It would be desirable to make calculations for the straight-line aircraft path to keep small changes for the angle between the wind direction and flight path direction. The aircraft experiment under con-

sideration is exactly fit for this purpose. At the same time there is certainly some distortions in the calculated results of the wind gradient during aircraft 'dip', its turnaround and also at the much more smooth ascent and descent stages.

Figures 4d and **4f** show that a significant correlation between the absolute wind gradient and small charged particles concentration is observed practically during all time of flight. The indicator of the charge particles is CN concentration which in an agreement with the equation (17) deduced from the Poisson equation (16).

That the Lorentz force affects first of all the charged particles with high mobility, these particles are ion clusters and small charged particles. It is just the wind which causes both not very large 'outbursts' of the small charged particles concentration and more significant variations of their concentration. A significant correlation between the inverse values of ozone content and CN concentration is evident (**Figure 4f** and **4h**).

The main ozone concentration rise (**Figure 4e**, ~5300 s and ~6800 s of the flight) is registered at the maximal wind velocity (**Figure 4c**) and close to zero absolute wind velocity gradient. In fact, the wind 'expands' the charged particles which are destruct ozone on both sides of the Polar vortex (**Figure 3b**).

An important conclusion from our analysis is the fact that ozone destruction takes part not only in the polar zone but also in the midlatitudes (compare **Figure 4f** and **Figure 4h**). Note that chlorine mechanism contribution exists only inside the cold vortex. The comparison of **Figure 4g** and **Figure 4h** shows also a rather high correlation between the chlorine monoxide concentration and inverse values of ozone content. Still this correlation is violated even inside the vortex during aircraft 'dip' (~61500 s) and also outside the vortex where the chlorine monoxide is almost absent (**Figures 4a, 4c**, **4g** and **4h**).

At the same time the anticorrelation between the CN and ozone concentrations (**Figure 4f** and **4h**) is observed throughout the whole aircraft flight and, from our point of view has probably a global character. During the aircraft 'dip', ascend, and landing the ozone destruction increases due to the small charged particle concentration rise in the upper troposphere. The tropospheric contribution to ozone destruction, as it was considered earlier, was found also in the balloon experiments (**Figure 1f** and **1h**). Chlorine monoxide is absent in the troposphere as well as outside the Polar vortex, and consequently, the chlorine mechanism does not participate in the ozone destruction in these areas.

It should be noted that the ozone depression formation in springtime in midlatutudes can be more hazardous for population that the ozone depression inside the Polar vortex in high latitudes because of the relatively less solar zenith angles and higher population density. The spring ozone concentration decrease is observed in reality but it is explained mainly by the ozone dynamics.

3.5 The role of the chlorine mechanism in the ozone depression formation

Let us consider the features of chlorine monoxide and ozone concentration variations in the polar lower stratosphere obtained from satellite observations.

Figure 5a–5d show spatial and temporal variations of ClO and O_3 concentrations for the winter and spring conditions for the Arctic, 2004/2005, and Antarctic, 2005, according to satellite data (Aura MLS) [2]. **Figure 5a** and **5b** give the winter and spring variations of ClO and O_3 contents in the air for various potential temperatures (altitudes) averaged for the region inside the Polar vortex (in the frames of the scaled contour of potential vorticity 1.6×10^{-4} s^{-1}). **Figure 5c** and **5d** show similar values for the Antarctic in the frames of the scaled contour of potential vorticity 1.4×10^{-4} s^{-1}.

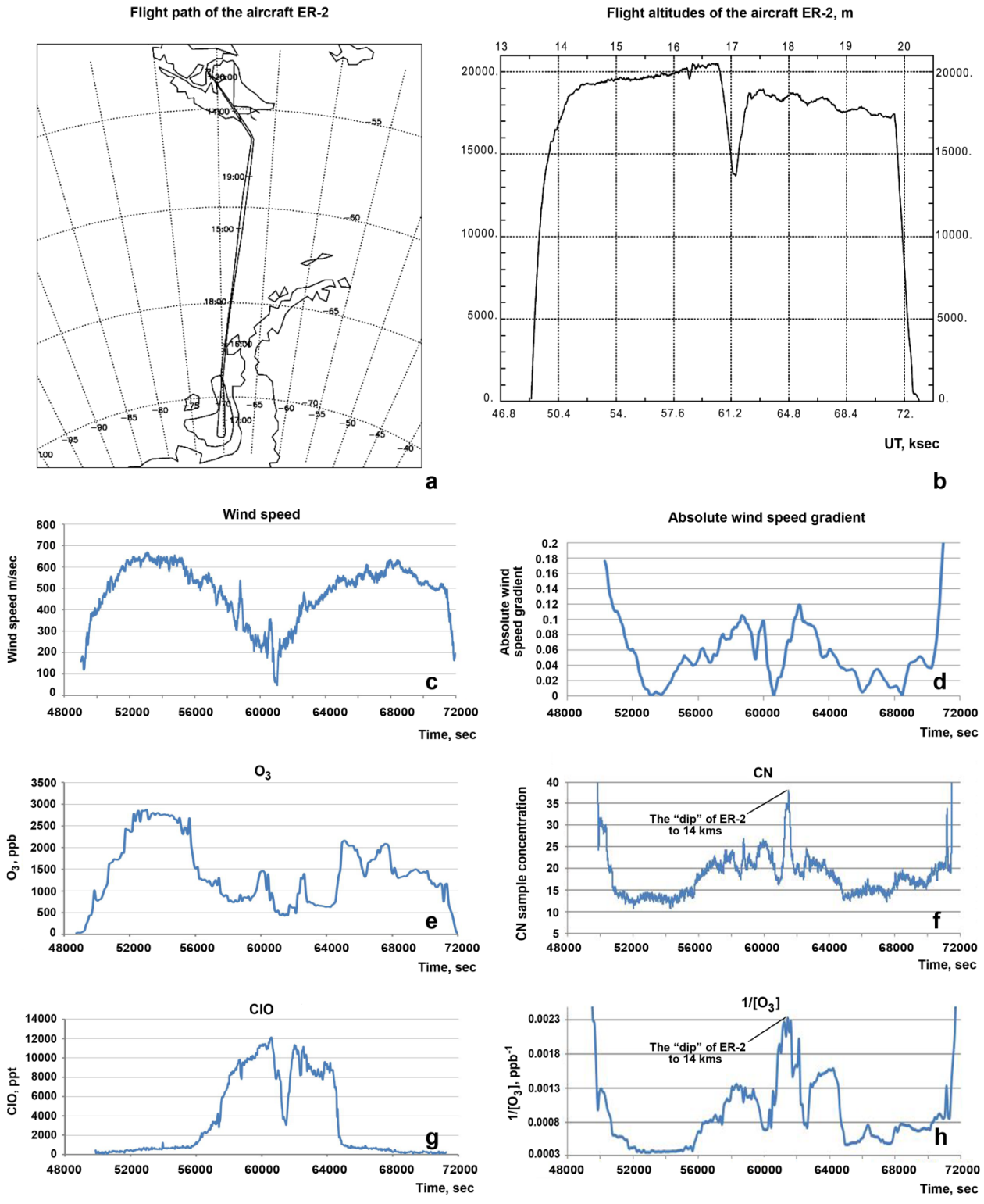

Figure 4. The results of measurements obtained from the ER-2 aircraft on September 22, 1987 in the AAOE-87 experiment. (**a**) flight path; (**b**) altitudes of the ER-2 flights; (**c**) wind speed; (**d**) absolute wind speed gradient (averaging for 500 sec); (**e**) mixing ratio of ozone (ppb); (**f**) CN concentration from the sampling; (**g**) mixing ratio of chlorine monoxide (ppt); (**h**) values of inverse ratio of ozone concentration (ppb^{-1}).

Source: AAOE-87 [30].

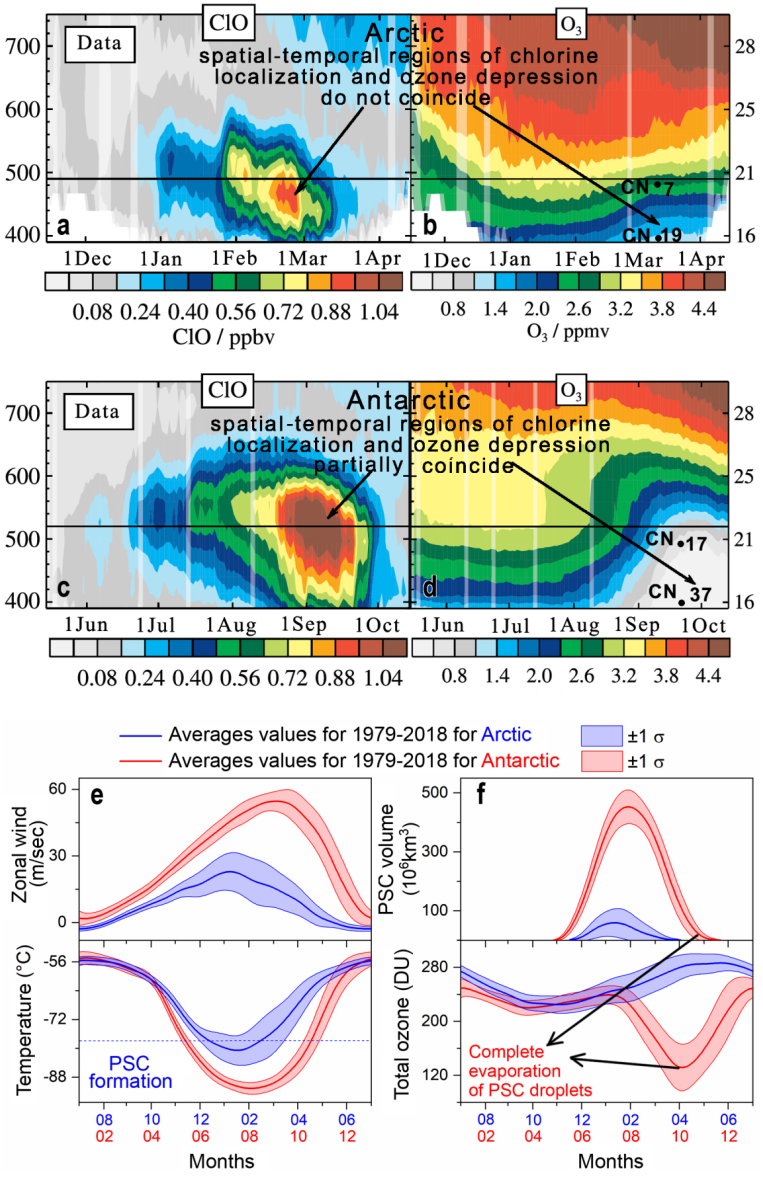

Figure 5. Spatial and temporal variations of ClO and O_3 for winter and spring conditions in **(a,b)** Arctic (2004/2005) and **(c,d)** Antarctic (2005) in the frames of the indicated scaled contour of potential vorticity (see text) based on satellite data. (**e**) Annual variation of the zonal wind velocity at 60º N/S and that one of minimal temperature in the lat. range 50º–90º N/S at the 50 hPa pressure level and (**f**) mean values of PSC volume and minimum of TOC for latitudes 40º–90º N/S for the period 1979–2018 with standard deviation ±1σ.

Sources: Santee et al [2, 3]

The spatial and temporal regions of the ClO localization and ozone depression in the Arctic do not coincide, while the same regions in Antarctica coincide only partially. The Arctic ozone depression began to develop in late February and early March at altitudes of ~16–18 km, reaching a maximum around on the first day of April and persisting thereafter. On the contrary, ClO decreases in this period and practically disappears by mid-March at all altitudes. The ozone depression maximum is situated much lower than the maximal chlorine monoxide concentration. The Antarctic ozone depression develops at ~16–25 km altitude range and its maximum is registered approximately on the first day of October and exists at least till mid-October, while ClO begins to decrease since the middle of September and disappears by the end of September. The ozone depression maxi-

mum is also displaced significantly relative to the maximum chlorine monoxide concentration(Figures 5a–d).

Such discrepancy between the regions of the chlorine monoxide localization and ozone depression can be related to the sinking of cold ozone-depleted air. But in our opinion, this is relatively unlikely. Analysis shows that in spring the lower stratosphere begins to warm up.

According to polar station McMurdo (78ºS) data, a significant temperature rise in the Antarctic is observed usually in September and especially in October inside the Polar vortex at altitudes 15–25 km [29] (**Figure 1j**). This altitudinal range corresponds to the localization of the chlorine monoxide in **Figure 5c**. Therefore, in springtime, the lifting of air more probably takes place due to its heating rather than its sinking. Taking into account the significant increase in temperature during the period under consideration also in the Arctic (**Figure 5e**), we conclude that the ozone depression formation in the lower stratosphere and upper troposphere at the altitudes below ~18 km in the Arctic and Antarctic is hardly probable (**Figure 5b** and **5d**).

Note that measurements shown in **Figure 5a–5d** are limited from below by the altitude of about 16 km. However, the balloon (**Figure 1**) and aircraft (**Figure 4**) measurements show that ozone depression can exist at lower altitudes in the upper troposphere. For this contribution to the ozone depression formation, we gave the name a 'tropospheric' one though this name is rather conventional because it may include also a certain altitudinal range in the lower polar stratosphere.

We state that not only a stratospheric contribution to the ozone depression formation but also a tropospheric one explains the specific features of the process of the ozone depression formation in the polar atmosphere.

The analysis of **Figure 1j** shows that in the polar atmosphere during seasonal change from winter to summer at altitudes ~15–17 km the temperature rise is much slower than that at the 20–25 km altitudinal range. This means that the PSC droplet's evaporation on the lower border lags in time relative to the droplet's evaporation on the upper border. **Figures 5a** and **5c** show that chlorine monoxide ClO sinks by the end of winter—beginning of spring following about the same time of the sinking of the upper layer of the CN increased concentration (**Figure 1i**).

At the same time, the development of the ozone depression at the altitude ~16 km and below by the middle of spring is related probably to the late temperature rise and growth of the charged CN concentration and subsequently with the time lagging of ozone depression formation in this region. It is important that in the process of the droplets evaporation, the surface potential of the charged particles increases significantly, and brings the O_3 molecules and molecules of other compounds to destruction during their collision with the charged particles.

The analysis of **Figure 5a–5d** showed that in Arctic a tropospheric contribution is observed mainly while in Antarctic both contributions are present: Stratospheric and tropospheric ones. That is why the spatial and temporal regions of the ClO localization and ozone depression do not coincide in fact in Arctic while in Antarctic they coincide only partially.

Figure 5b and **5d** show CN concentration variations during ER-2 aircraft 'dip' in the Arctic experiment performed on March 11, 2000 [5,35] and in the Antarctic experiment which was considered above and performed on September 22, 1987 [30] (**Figure 4f**). The flight dates were synchronized with the dates given in **Figure 5b** and **5d**. The CN concentration rise during flight descent corresponds to the decrease of the ozone content in the atmosphere in both experiments. This fact, in our opinion, indicates the direct relation between ozone destruction and small particle concentration.

Note, that approximately in October, 1 multi-year minimum of the total ozone content is registered in the 40°–90°S latitudinal belt [3]. **Figure 5e** and **5f** show the means for temperature, zonal wind, PSC volume and total ozone content for almost 40 years at the pressure level of 50 hPa (about 20 km altitude) in the Arctic and Antarctic [3].

The maximum development of the ozone depression on October, 1 corresponds to the practically

total disappearance of PSC (**Figure 5d** and **5f**). According to our analysis, this moment of complete evaporation of charged droplets approximately corresponds to the moment of maximum formation of the small charged particles (**Figure 2**) at which the most intense ozone destruction occurs. This supports our hypothesis that, in reality the main mechanism of the ozone depression formation is the evaporation and fragmentation of the electrified droplets of the polar clouds in springtime.

At the same time, as can be seen in **Figure 5f**, based on measurements of the total ozone content in the spring in the Arctic, no ozone minimum is detected at all. However, this does not mean that the ozone depression does not emerge in spring. It can appear in some altitudinal range. According to satellite measurements, a rather strong depression appears at the altitudes below 18 km (**Figure 5b**). The aircraft and balloon experiments reveal that the depression can arise even in the upper troposphere (**Figure 1** and **4**). The maximum of the ozone depression for these altitudes is registered approximately April, 1 (**Figure 5b**).

Figure 5e shows that by this date the Arctic Polar vortex decreases strongly and its collapse and also the temperature rise (**Figure 1j**) go 'top-down'. Probably, the ozone concentration rise at altitudes above 20 km is caused by the propagation of air which is enriched by ozone from the mid-latitudes due to the decrease and collapse of the Polar vortex at those altitudes (**Figure 5b**). Thus, a peculiar kind of ozone compensation takes place at other altitudes in the Arctic region. A similar compensation in the Antarctic is less typical (**Figure 5d**), probably because of lower temperatures of the polar atmosphere and the greater vortex dynamical stability (**Figure 5e**).

At the same time, at least in some cases of the strongest depression in Arctic, the ozone depression can be registered from the total ozone content (TOC) measurements, especially outside the vortex where effect of the vortex collapse on the total ozone content must be less significant.

An example of the development of such ozone depression in the Northern hemisphere in Russia in the middle latitudes is given in Dorokhov's study.[31]. **Figure 6** shows the annual cycle of the total ozone content (TOC) in 2011 measured by the Brewer spectrophotometer in the cities of Tomsk (56.5°N, 85.0°E) and Obninsk (55.1°N, 36.3°E) [31]. The average monthly values of TOC in Tomsk and Obninsk are shown by the green solid line. The blue dashed lines show the standard deviation of ± 2 σ from the mean climatic value. The black ovals mark the period of the unusually low TOC in Tomsk and Obninsk in the spring of 2011.

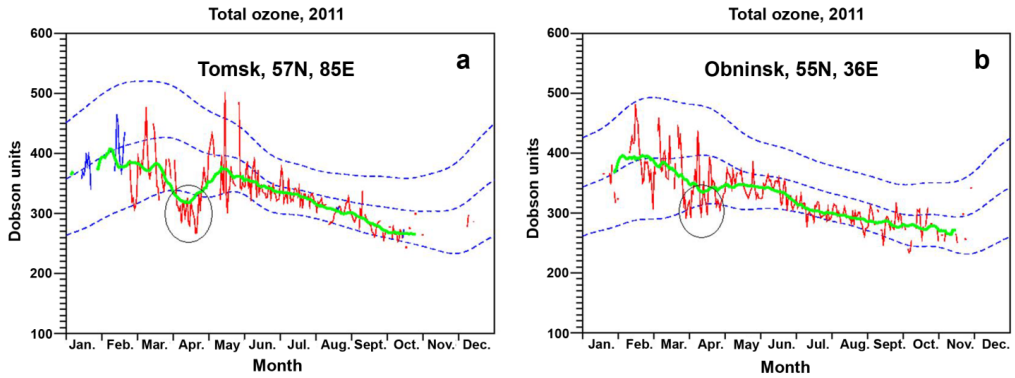

Figure 6. Total ozone content was measured by (**a**) the Brewer MKIV S/N 049 in Tomsk in 2011 and (**b**) the Brewer MKII S/N 044 in Obninsk. A dip can be observed in the Tomsk data in early April 2011 when the ozone-depleted vortex moved towards central Siberia. The green line shows the monthly averaged mean values of total ozone content in Tomsk and Obninsk in 2011. The blue dotted lines show ± 2 sigma standard deviation from the climatic mean. The black ovals show the time period of unusually low total ozone in Tomsk and Obninsk in spring 2011.

Source: Dorokhov et al [31].

Figure 6 shows that the springtime ozone depression in the Northern Hemisphere is steadily observed in the background of the natural variations and spreads far over the polar region. Indeed, the 2011 ozone depression was one of the strongest, which was observed since 2000. Our analysis shows that a possible mechanism which enhances the ozone depression development in mid-latitudes is the effect of the Earth's magnetic outside the Polar vortex on the motion of the charged particles which are transported by the Lorentz force toward the middle latitudes (**Figures 3a** and **4**).

Thus, according to our analysis, the chlorine mechanism's contribution to the ozone destruction in the Arctic is insignificant, though sometimes it is possible that the role of this mechanism can be enhanced. Still, the main mechanism is direct ozone destruction with the participation of ion clusters and small charged particles below ~18 km.

At the same time, in the Antarctic, as it was mentioned above both stratospheric and tropospheric contributions into the ozone depression formation exist. However, the combined effect of these mechanisms can appear only inside the cold Polar vortex at the lower stratosphere altitudes. It is difficult to separate them, based only on the experiments considered above, but in future investigation, it is necessary to specify the contribution of each mechanism into ozone depression formation.

4. Conclusions

Based on the experimental data analysis and theoretical estimations, a possibility of the existence of a new mechanism of the ozone destruction with the participation of ion clusters and small charged particles in the polar atmosphere was shown. A proposed mechanism of the ozone depression formation expects the accumulation of the charge cloud particles in the upper troposphere and lower stratosphere preferentially in the cold wintertime and the transformation of the large charged particles into the small ones in spring due to the process of evaporation and fragmentation of the charged cloud particles. Moreover, due to this avalanche-like process of the size degradation of the charge particles practically all charge which was accumulated on the large charge particles can be transformed into ion clusters with a sufficient temperature rise [36–41].

Such process of charged particle evaporation can be observed probably at various cloud levels, from the troposphere one to even the mesosphere level and probably has a global character. However, it is important that the concentration of the charge which is accumulated on the cloud particles and then 'releases' as small charge particles and ion clusters in a process of the evaporation and fragmentations of droplets should be sufficient for the ozone depression formation.

The effective mechanisms of the charge accumulation on the particles are the Global Electric Circuit operation and, as it was revealed, the Earth's magnetic field which affects the charged particles' motion inside the Polar vortex. The main factor of those mechanisms' effectivity is the charge separation by their polarity and accumulation of the unipolar charge on the cloud particles. In this case, the influence of recombination of particles with different signs of charge is reduced strongly and the concentration of accumulated charge increases significantly.

Note, that charge separation takes place in different conditions. If the unipolar charge is accumulated on the upper and lower flat cloud boundaries due to GEC operation, then the vertical component of the magnetic field separates moving charges on the different sides of the vortex. Both mechanisms of charge accumulation can exert influence on each other; they can either be stronger or weaker in certain regions of the polar atmosphere.

Inside the vortex, both mechanisms expose a stronger effect on the upper cloud boundary in the lower stratosphere, where a positive charge is accumulated, and both become weaker at the lower cloud boundary. Outside the vortex at the same time, a mutual enhancement of those mechanisms occurs at the altitudes of the upper troposphere on the lower clouds boundary, where a negative charge is accumulated, and their weakening on the altitudes of

the lower stratosphere. Thus, the main stratospheric contribution is centred inside the vortex in the lower stratosphere. However, a tropospheric contribution also exists, and its effect, according to our analysis, should show itself most strongly outside the vortex. Moreover, the springtime depression can spread to mid-latitudes. More relevant results of this work need further detailed study and proof of our concept.

Author Contributions

All authors of this research under the guidance of Dr. Belikov contributed to the final version of the manuscript.

Conflict of Interest

The authors declare that there is no conflict of interest.

Data Availability Statement

The data that support the findings of this study are openly available in the references [2–5, 27–29,30,31,37] of this paper.

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