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ATMOSPHERIC RADIATION, OPTICAL WEATHER, AND CLIMATE

Changes in Surface Ozone Concentration during Precipitation

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Abstract—Changes in ozone concentration during precipitation are studied using data of ozone monitoring in the surface air layer in Tomsk and at Karadag. It is found that these changes can be either positive or negative. The largest jumps in ozone content are observed during frontal precipitations. During air mass precipitation, the sign and magnitude of the changes are determined by the diurnal behavior of ozone concentration. Analysis showed that the increase in the ozone concentration during precipitation and ozone increase in the diurnal behavior are time-coincident in 59% of cases in Tomsk and in 63% of cases at the Karadag. The decreasing wave of ozone concentration in the diurnal behavior coincides in time with ozone decrease during precipitation even more often, in 85% of cases in Tomsk and in 79% of cases at Karadag. Based on data of aircraft sensing, it is shown that ozone descent from the boundary air layer occurs in a number cases when the temperature stratification during precipitation changes to neutral.

Keywords: atmosphere, gas, air, washout, ozone, precipitation, admixtures, removal
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INTRODUCTION

Ozone, as one of trace gas admixtures of air, is not emitted by natural and anthropogenic sources, but rather is formed directly in the atmosphere via photochemical processes from precursor gases. The variety of precursor gases and the complexity of ozone production determine the fundamental value of ozone studies. At the same time, their practical necessity stems from ozone properties and, in particular, from the fact that ozone is the strongest oxidant, having a destructive effect on environmental structures; ozone in large concentrations is a poison, intoxicating the biosphere; and ozone is a radiatively active gas whose contribution to the greenhouse effect is ranked fourth in significance.

Ozone is one of the most studied air admixtures, with thousands of works published on the subject. Nonetheless, many fundamental issues of ozone formation and transformation in the atmosphere are still poorly or even insufficiently well studied.

In overview [1], compiled by an international group of scientists, it is noted that variations in ozone concentration in the troposphere are determined mainly by ozone transport from the stratosphere and photochemical ozone production from precursor gases, depending on their composition and concentration, influx of solar radiation, and meteorological factors. Precipitation is one of the poorly studied meteorological factors, influencing the surface ozone concentration.

The process of interaction between ozone and precipitation was considered theoretically in work [2], by analyzing two variants: adsorption of gas molecules onto the particle surface and chemical reaction with the particle surface. No evidence on how these theories were verified in the literature were found; however, there are indirect indications that precipitation and ozone can be interrelated. For instance, in a number of works it is shown that ozone concentration decreases by a factor of two in the monsoon period [3–6]. Statistical prognostic ozone models incorporate precipitation as a predictor [7, 8]. The largest ozone concentrations are observed at minimal precipitation amounts [9]. Aircraft measurements in clouds [10] indicate that ozone content decreases a little (by 3–4 ppb) along aircraft transit across the cloud. Our measurements suggest that ozone concentration in clouds decreases on the average by 15 ppb [11]. The authors of works [12, 13] obtained estimates intermediate to [10, 11]. Also, there are publications which show that precipitation weakly influences the ozone concentration [14, 15].

Thus, at present there is no consensus about how precipitation influences the surface ozone concentration; therefore, this work is aimed to analyze whether or not the O_3 content changes during precipitation.

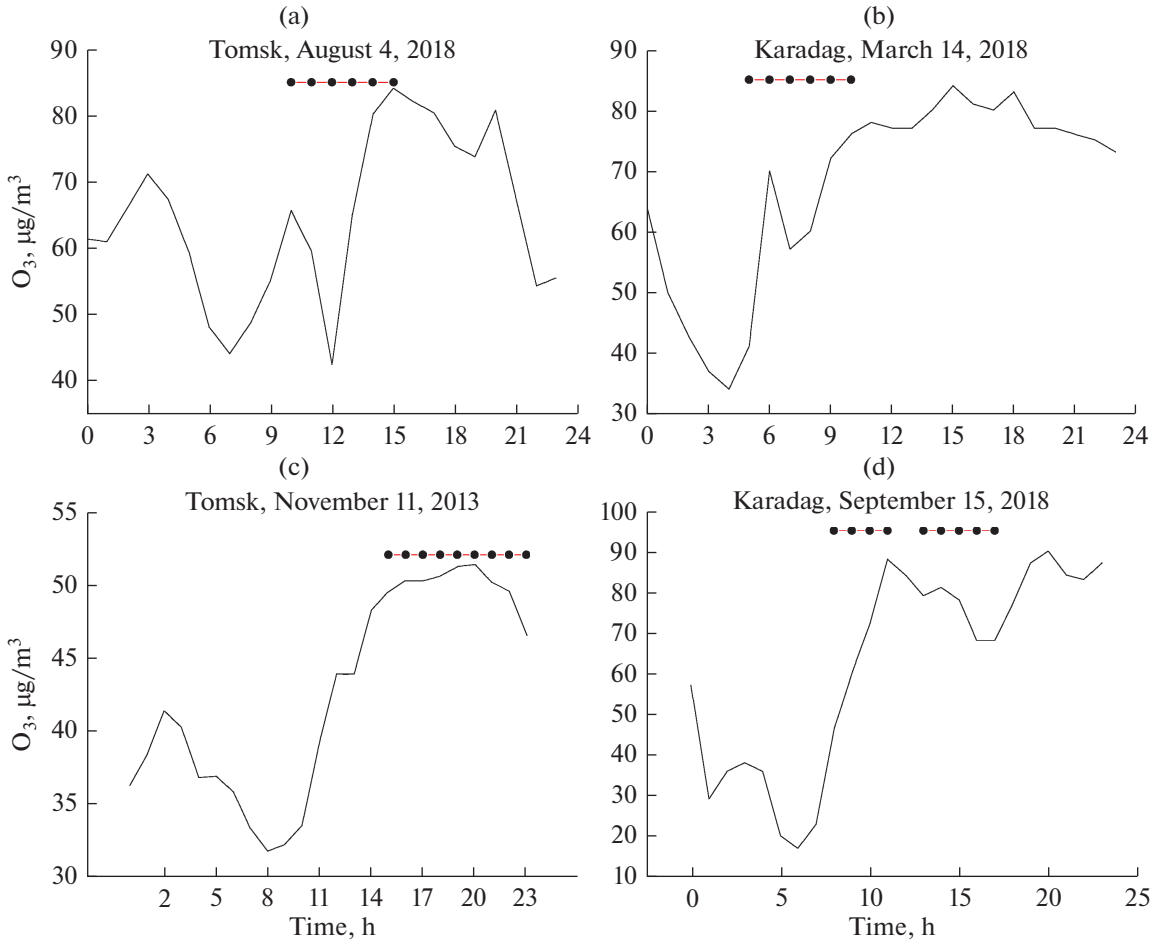

Fig. 1. Changes in surface ozone concentration (a, c) in Tomsk and (b, d) at Karadag; horizontal line with circles indicates the periods with precipitation.

DATA AND STUDY REGIONS

The study was performed using data on monitoring air composition in the region of Tomsk, Akademgorodok, at the TOR station, the location and the current state of the measurement complex which were described previously in [16], and data from the station for background ecologic monitoring (SBEM) at Karadag Scientific Station, Nature Reserve of the Russian Academy of Sciences (44°55′ N, 35°14′ E, 180 m ASL). At the SBEM, ozone was measured by the chemiluminescent method using 3.02P-A automatic gas analyzer (OPTEK, Russia) until March 20, 2013. Since then to the present, monitoring has been performed by the optical method using an APOA-370 gas analyzer (HORIBA, Japan). Samples were taken using Teflon pipes at altitude of 2 m above the Earth's surface. For analysis, we employed synoptic data obtained at the WS-600 meteorological station (Germany).

In this work we used measurements for 2013 and 2018 when maximum and minimum annual average ozone contents over last decade were observed in Tomsk. Data for these same years are also taken for Karadag to synchronize the comparison. The purpose of the paper was to clarify the nature of the physical interaction between ozone and precipitation; therefore, the choice of the period of time was not critical because the mechanism should be manifested independent of the period and geographic positions of observation sites.

ANALYSIS AND DISCUSSION

For study, we selected days with precipitation and analyzed changes in surface ozone concentration in periods with precipitation. It was found that, independent of the precipitation type, ozone content can both decrease and increase. Figure 1 presents specific cases for Tomsk and Karadag.

From Figs. 1a and 1b it can be seen that surface ozone concentration decreased by 24 and 12 μg/m^3 in Tomsk and at Karadag, respectively, during precipitation. Figure 1c shows that ozone concentration increased by 6 μg/m^3 during precipitation in Tomsk.

At Karadag on September 15, 2018, (Fig. 1d) the rain was with an interruption, so that ozone concentration increased by 28 μg/m^3 during first rainfall episode and decreased by 16 μg/m^3 during second episode.

Analysis showed that the intensity, type, and duration of precipitation do not influence the changes in ozone concentration (Fig. 2).

Figure 2a was plotted using data for May 16 and 29, June 3, and September 3, 2013, for Tomsk and for February 12, March 3, July 28, and October 2, 2018, for Karadag. It can be seen that ozone concentration can change by 1.2 and 21.5 μg/m^3 at the precipitation rates of 0.6 and 0.5 mm/h, respectively, and by 3.4 and 25.8 μg/m^3 at the precipitation rates of 8.5 and 8.4 mm/h, respectively. A similar nonunique pattern is also recorded at Karadag: ozone concentration can change by 2.0 and 22.0 μg/m^3 when the precipitation rate is small (0.4 and 0.2 mm/h); and concentration may change both by 3.0 and 14.0 μg/m^3 after the precipitation rate increases to 4.4 and 3.7 mm/h. Thus, no unique interrelation between changes in ozone concentration and precipitation rate is evident in Fig. 2a.

We selected the cases with approximately identical precipitation intensities in Tomsk (Fig. 2b) to compare the changes in ozone concentration during rainfall and snowfall. Snowfall is rare in Karadag, for which this procedure could not be performed. It was shown that ozone concentration depends uniquely neither on precipitation type nor intensity. The difference in ozone concentration between rainfall and snowfall can be explained by specific feature of the annual behavior of ozone, the maximal (minimal) content of which is usually recorded in spring–summer (late fall–early winter) [17].

No dependence between variations of ozone content in the surface air layer and precipitation duration can also be found in Fig. 2c. This figure was plotted using data for April 23, June 11, August 25, and November 2, 2018, for Tomsk and for May 8, July 6, September 15, and October 2, 2018, for Karadag. We note that changes in ozone concentration during precipitation are somewhat stronger at Karadag. This may be because of geographic features of the regions.

In Tomsk, 115 days with precipitation were recorded in 2013 and 88 days in 2018 (Table 1). At Karadag, the number of days with precipitation is smaller: 54 days in 2013 and 57 days in 2018. Table 1 presents the number of cases with precipitation, during which the ozone concentration either increased or decreased.

From data in Table 1, it follows that a decrease in ozone concentration is observed more frequently than ozone increase during precipitation in both Tomsk and Karadag. Presumably, ozone removal from the atmosphere is more probable during precipitation.

This asymmetry will be addressed below. Here, we note that ozone is poorly soluble in water and, as such,

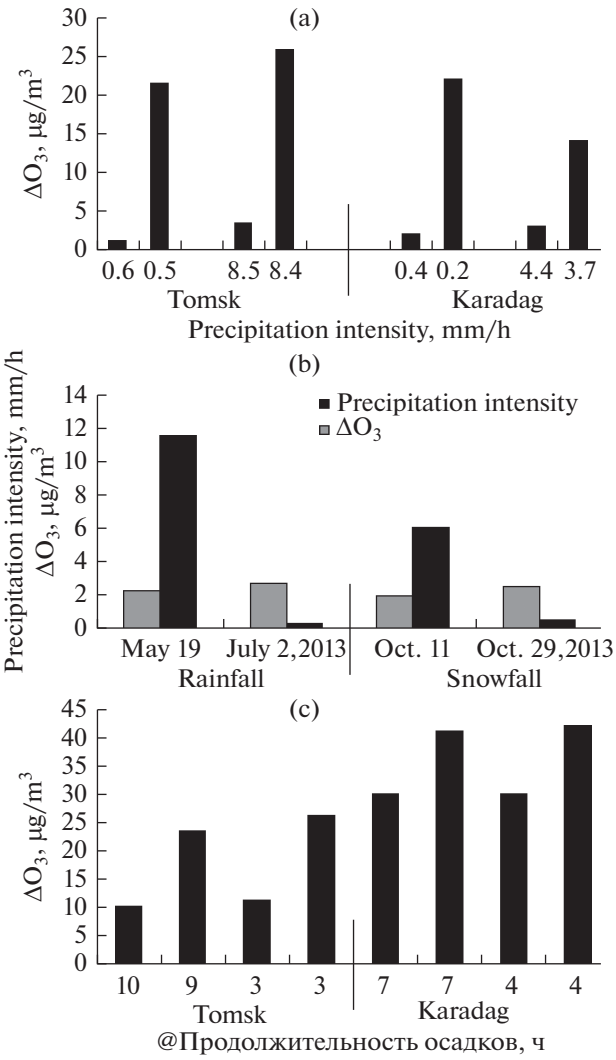

Fig. 2. Changes in the surface ozone concentration in Tomsk and at Karadag as functions of precipitation intensity, type, and duration.

is barely washed out [18, 19]. Other mechanisms seem to be responsible for changes in zone concentration.

Analysis of the dynamics of ozone concentration during frontal passages [20] shows that onset of precipitation can lead to both decrease and increase in ozone concentration by 30% on average. This effect

Table 1. Frequencies of occurrence of cases with increasing and decreasing ozone concentrations in Tomsk and at Karadag

Region	Year	Increase	Decrease	Total
Tomsk	2013	45	70	115
	2018	31	57	88
Karadag	2013	19	35	54
	2018	24	33	57

Table 2. Frequencies of occurrence of increasing and decreasing ozone concentrations during precipitation on increasing and decreasing waves of concentration in diurnal behavior in Tomsk and at Karadag

Region	Increasing O_3 concentration, number of cases, %	Decreasing O_3 concentration, number of cases, %	Total number of cases, 100%
Increasing wave of O_3 concentration in diurnal behavior			
Tomsk	17 (59)	12 (41)	29
Karadag	12 (63)	7 (37)	19
Decreasing wave of O_3 concentration in diurnal behavior			
Tomsk	7 (15)	39 (85)	46
Karadag	4 (21)	15 (79)	19

was found by other researchers [21–24]. It is noteworthy that, as shown in [25], ozone content inside an air mass is quite homogeneous; and ozone concentration jumps after a change of air mass. Whether the concentration increases or decreases depends on which air mass comes in place of the previous air mass at the observation site. Analysis [25] revealed that in the region of Tomsk the largest (smallest) ozone concentration is observed in the tropical (Arctic) air mass; therefore, the concentration decreases when a midlatitude air mass changes to an Arctic air mass, and vice versa. It is important to stress that this pattern does not always hold in the real atmosphere. As noted earlier by S. P. Khromov in [26], fronts may strongly differ, and the field of vertical motions may be differently formed in them; therefore, there are deviations from this ideal pattern in 20–25% of cases due to specific features of dynamical processes in frontal zones [20, 27]. Hence, cases with frontal precipitation were excluded from consideration.

On the one hand, this worsened the total statistical representativeness. On the other hand, the asymmetry of the frequency of occurrence of increasing and decreasing surface ozone concentration during air mass precipitation did not change, indicating that other causes are responsible for changes in the O_3 concentration during precipitation. In our opinion, the diurnal behavior of the surface ozone concentration, which is well defined in both regions considered here, can be such a cause [28–30].

To test this hypothesis, all cases of air mass precipitation were divided into two groups. The members of the first (second) group were precipitation episodes, observed on an increasing (decreasing) wave of ozone concentration in the diurnal behavior. The authors of work [28] argue that the increasing wave of O_3 concentration in Tomsk is observed from 9:00 to 16:00 Local Time (LT) in the cold period and until 18:00 LT in the warm period; and a decreasing wave is observed from 17:00 to 8:00 LT in the cold period and from 19:00 to 8:00 LT in the warm period. The increasing (decreasing) wave at Karadag is observed from 9:00 to 18:00 LT (from 19:00 to 8:00 LT) [29, 30].

This division substantially changed the relationship between the frequencies of occurrence of increasing and decreasing ozone concentrations during precipitation, though not with the 100% success. This can be judged from data in Fig. 3.

In Figs. 3a and 3b it can be seen that ozone content decreased both in Tomsk and at Karadag during precipitation observed in a decreasing wave of ozone concentration in diurnal behavior. Figures 3c and 3d indicate that ozone concentration may also increase on this same decreasing wave in diurnal behavior.

Results of analysis for all cases are presented in Table 2. It should be noted that no division into years was performed because of low residual statistics.

Data of Table 2 demonstrate that ozone concentration increases during precipitation simultaneously with its growth in diurnal behavior in 59% of cases in Tomsk and in 63% of cases at Karadag. A decreasing wave of concentration in its diurnal behavior coincides in time with an ozone decrease during precipitation even more frequently: in 85% of cases in Tomsk and in 79% of cases in Karadag. Better results could be obtained through an individual selection and a shift of periods of decreasing and increasing O_3 concentration in the diurnal behavior; however, a formal approach was kept to avoid the inevitable effect of the subjective factor.

Thus, a factor determining the change in ozone concentration during the air mass precipitation is the period when the precipitation falls. If the precipitation occurs on the increasing wave of ozone concentration in the diurnal behavior, the ozone content also increases in the surface layer in most precipitation events. On the contrary, O_3 content mostly decreases if rainfall or snowfall coincides in time with the decreasing wave of ozone concentration in the diurnal behavior.

This result also helps to explain the asymmetry of the frequency of occurrence of cases with increasing

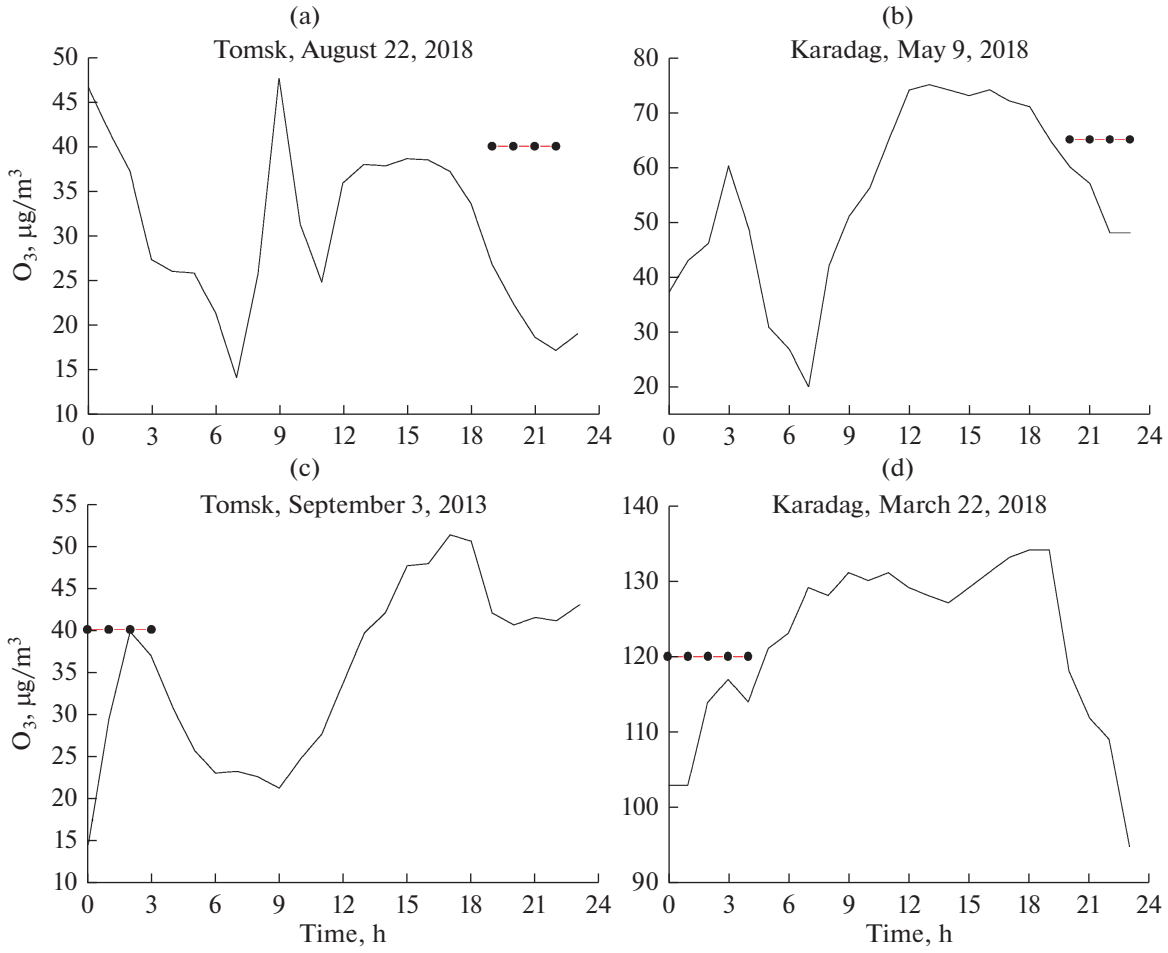

Fig. 3. Changes in surface ozone concentration (a, c) in Tomsk and (b, d) at Karadag during air mass precipitation (indicated by line with circles).

and decreasing ozone concentration during precipitation. It reflects the timing of precipitation relative to the diurnal ozone behavior. For instance, the period of increasing (decreasing) ozone concentration in the diurnal behavior is 7–9 h (13–15 h), depending on the time of the year or a region. Whereas the probability of precipitation is identical at any time of the day, the precipitation events on the decreasing wave of ozone concentration in the diurnal behavior will be much more frequent owing to a much longer duration of this period of time. This fact was just noted at the beginning of this paper.

Data, presented above, indicate that the variations in O_3 concentration during precipitation in 15–40% of cases occur due to some other reason than that described above. Most probably, this is because the maximum in the diurnal behavior of photochemical ozone production is not always observed near the Earth's surface. It was shown theoretically in [31, 32] that the maximum of photochemical O_3 production should be observed inside the boundary layer at the altitude reached by ozone-forming species transported from the surface. This was confirmed experimentally in [33–35].

In precipitation, a droplet or snowflake entrains an air parcel and delivers it to the Earth's surface [36]. Instead, air comes from the underlying layers. As a result of this dynamic process, concentrations of admixtures are aligned vertically, as the temperature stratification [37, 38]. In the course of this process, soluble gases and aerosol are scavenged from the atmosphere [39–41]. Insoluble gases are carried to either overlying or underlying areas. For instance, carbon oxide is shown [42] to enter the subcloud layer, and ozone enters the surface air, provided that the ozone concentration is higher in the boundary layer.

This will be shown by the example of aircraft sensing performed on the Mirny–Lensk–Bratsk route on September 8, 2006. We used the instrumentation as described in [43]. The synoptic situation in the period of the experiment is shown in Fig. 4; and the data on the vertical stratification are presented in Fig. 5.

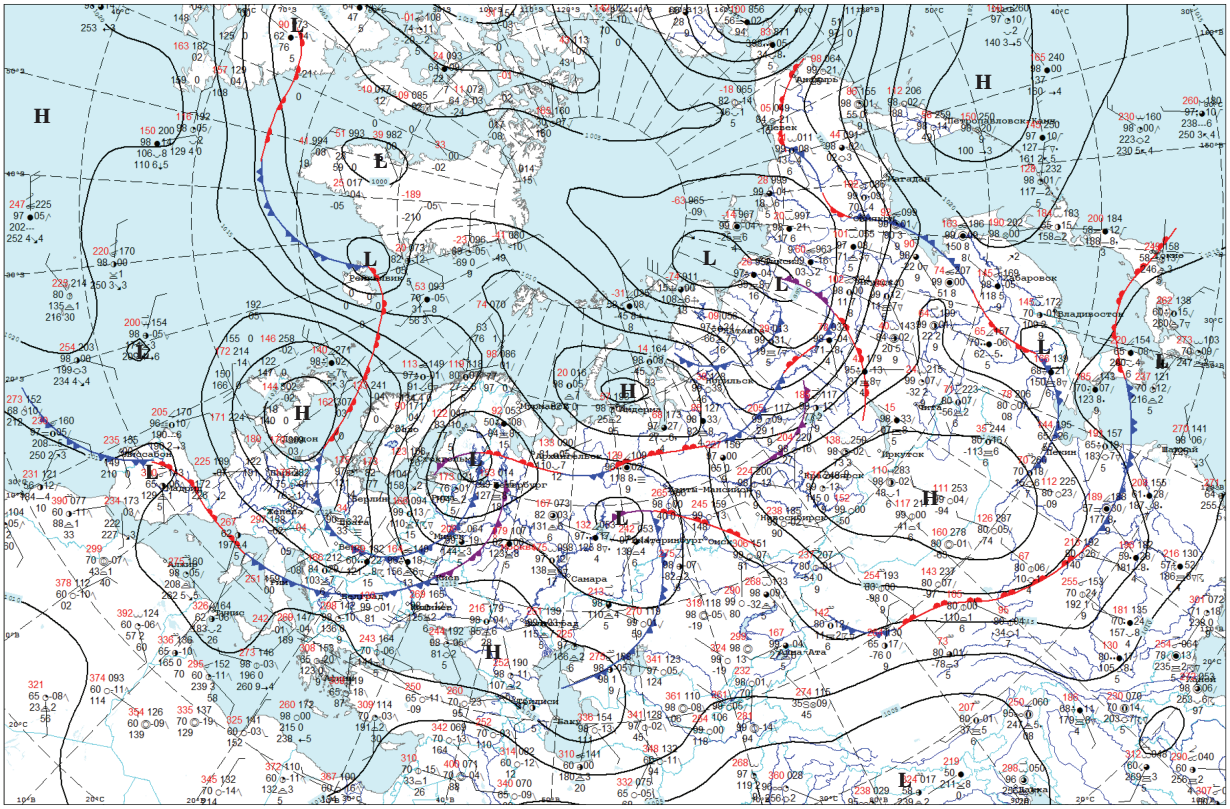

Fig. 4. Surface weather map on September 8, 2006 (12:00 UTC).

Aircraft took off in Mirny in a midlatitude air mass and landed in Lensk in an Arctic air mass before the approaching warm front. No precipitation was observed during landing. Drizzling rain began one hour before takeoff, so that the aircraft took off in moderate-intensity precipitation. The aircraft landed in Bratsk after crossing warm front.

The profiles of temperature and relative humidity in Fig. 5 reflect the character of their distribution at landing and takeoff sites.

In Fig. 5a, it is clearly seen that the temperature stratification of the atmosphere was unstable in the lower 400-m air layer during landing in Lensk and stable in the atmosphere above. During takeoff, the temperature stratification in the layer from 200 to 1000 m became neutral, consistent with results in works [37, 38]. The lower (upper) cloud edge was at the 850-m (about 3000-m) level, as can be seen in Fig. 5b.

Figure 6 shows how ozone concentration varied during this period. Ozone concentration in the atmosphere during landing in Lensk was lower than in the neighboring cities. Ozone concentration increased considerably during takeoff, both in the surface air and in the subcloud layer, so that a concentration maximum had formed at 1700-m level inside the cloud. Under the conditions of neutral temperature stratification and intense dynamic mixing due to precipitating droplets, ozone entered the surface air layer.

A similar process was also recorded on the territory of the Amazon [44]. As in our case, the first (second) episode was accompanied by an increase (a decrease) in ozone concentration.

CONCLUSIONS

Our study shows that atmospheric precipitation may be accompanied by either an increase or decrease in surface ozone concentration, independent of precipitation type, intensity, and duration, as well as of the region of measurements.

The largest changes are associated with frontal precipitation events and reflect a jump in ozone concentration during a change of air mass.

The sign of a change in ozone content during air mass precipitation depends on the time of the day when the precipitation falls. If precipitation occurs on the increasing wave of O_3 concentration in diurnal behavior, ozone content increases in most cases. On the contrary, if precipitation occurs on decreasing wave of O_3 concentration in diurnal behavior, ozone content decreases.

When stratification of the atmospheric boundary layer changes during precipitation, ozone concentra-

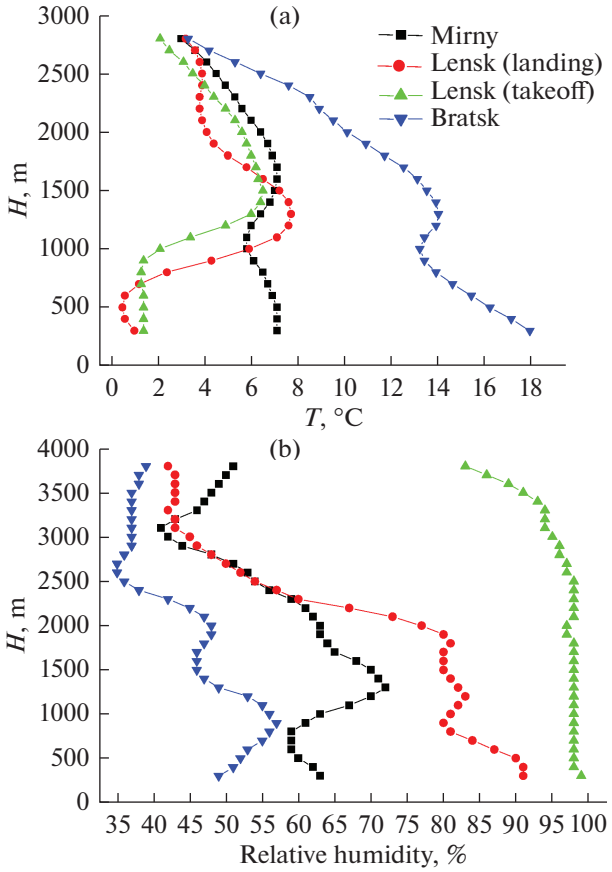

Fig. 5. Vertical distributions of (a) temperature and (b) relative humidity of air on September 8, 2006.

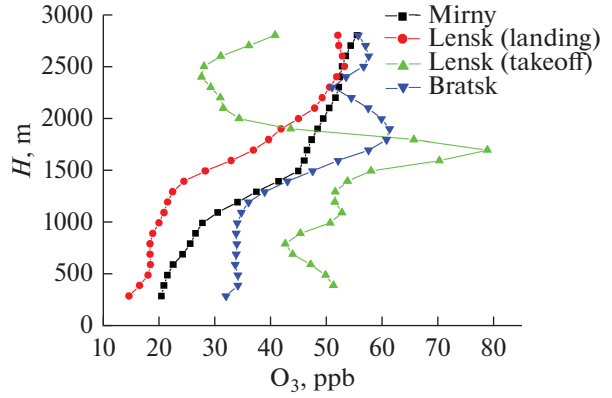

Fig. 6. Vertical distribution of ozone concentration on September 8, 2006.

tion can either increase or decrease, depending on where (at which altitude) the ozone content is greater.

No process of ozone washout by precipitation was found.

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CONFLICT OF INTEREST

The authors declare that they have no conflicts of interest.

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