Author's response

Case study of ozone anomalies over northern Russia in the 2015/2016 winter: Measurements and numerical modeling

5 By Yury M. Timofeyev, Sergei P. Smyshlyaev, Yana A. Virolainen, Alexander S. Garkusha, Alexander V. Polyakov, Maxim A. Motsakov, Ole Kirner

Abstract. Episodes of extremely low ozone columns were observed over the territory of Russia in the Arctic winter of 2015/2016 and the beginning of spring 2016. We compare total ozone column (TOC) obtained using different remote sensing techniques (satellite and ground-based observations) and results of numerical modelling over the territory of the

- 10 Urals and Siberia for the above period. We demonstrate that the provided monitoring systems (including new Russian Fourier- spectrometer IKFS-2) and modern 3-dimensional models are able to capture the observed TOC anomalies. However, the results of observations and modelling show discrepancies of up to 20-30% in TOC measurements. Analysis of the role of chemical and dynamical processes demonstrates that observed short-term TOC variability is not a result of local photochemical loss initiated by heterogeneous halogen activation on particles of polar stratospheric clouds that formed under
- 15 low temperatures in the mid-winter.

Reply to reviewer 1

Dear Referee,

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Thank you for your comments on the paper and constructive recommendations. We have tried to follow your suggestions and have utilized most of them. Following we mention how the manuscript has been changed according to your comments.

25 General comments:

For example, Section 3 on the "Comparison of total ozone column measurements and numerical modeling", one of the papers' essential sections, does not discuss at all the rationale of the use of two different models. Moreover, the two different models are forced (or nudged) with different data sets. Both of these facts introduce errors and

discrepancies in the comparisons, which are not discussed at all. Moreover, there is no information on the comparison between the two models and the comparison between the two forcing data sets. Please correct also the figures, and clarify the Figure Captions. Figure 2 and Figure 3 contain solid lines with different shades of gray, and it can be hard to distinguish between them.

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ACCEPTED

- 1. In Section 3, the motivation for using two different models is added, consisting in an attempt to assess the impact of the interactive interaction of the chemical and dynamical processes (the
- 10 EMAC model) with a re-analysis data nudging against the background of using re-analysis data in the RSHU model. In addition, the models have different spatial resolution, which makes it possible to estimate the effect of model resolution on the comparison with the observations related to the local points.
 - 2. An additional numerical experiment using the ERA-INTERIM reanalysis data was performed
 - with the RSHU model in order to compare the effect of different meteorological data on the comparison of the results of numerical modeling and local observations.
 - 3. At the end of section 3, the results of a comparison of numerical modeling and observations, as well as comparisons between models with different meteorological data, are expanded.
 - 4. Figures 2 and 3 are made in color, and the RSHU model simulation results with the EPA-
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INTERIM data are added in Figure 3.

Specific comments.

1. Page 2, lines 10-11 end elsewhere in the paper: "TOC depletion: : :" Short term episodes of low ozone values are better described by "ozone loss" rather than depletion, a term which implies a longer-term decay with

25 significant duration

2. Page 3, line 10: "three long periods of essential deviations of the average daily TOC: : :" I do not understand what is meant here. Please clarify

3. Page 3, line 10:": : : from average long-term values: : :" What is the time period for this climatological average that you refer to?

4. Page 3, line 13:"of average values (191-257,:::)" Please clarify what is this range of values referring to?
5. Page 3, line 18. Please indicate the location of the stations in Figure 1, and/or in Figures 4 and 5. It will be extremely helpful for the reader's understanding.

6. Page 3, line 24 "including observed ozone depletion". Do you mean here the ozone depletion in general (e.g. Northern hemisphere), or short-term ozone loss? Please clarify.

35 7. Page 3, line 30: "...during periods of ozone depletion" Again, does this refer to a general longer term behavior, or the short period examined here?

8. Page 3, line 31-32:" This then described the mean state : : :" I do not understand the meaning of this sentence 9. Also in line 32-33, "to improve: : : with the appearance of strong ozone anomalies". Do you mean inclusion of ozone anomalies? It is not clear at all.

10. Page 4, "Comparison of total ozone column : : :" Section Please see my general comment in the beginning

5 11. .. Page 4, line 25: Figure 3. It is not clear which line is for each model. I can see (and print) two gray lines. Please correct.(also in Fig. 2)

12. Page 5, lines 10 and below: What is printed in the lower panels of Figure 5? The text refers to MERRA, but the figure caption refers to model output.

10 ACCEPTED

1. Page 2, lines 10-11 and elsewhere. The depletion of ozone is everywhere replaced by ozone loss.

2. Page 3, line 10-14. Clarification has been done.

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3. Page 3, line 10. Climatological period is specified to be from 1979 to 2017.

4. Page 3, line 13. Clarification for the ranges of values has been done.

- 5. Page 3, line 18. Location of stations has been indicated at the fig.2.
 - 6. Page 3, line 24. Clarification for short-term ozone loss have been done.
 - 7. Page 3, line 30. Short-term period of ozone loss is clarified.

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- 8. Page 3, line 31-32. The sentence has been modified.
- 9. Page 3, line 32-33. The sentence has been corrected.
- 30 10. Page 4. Section 3 has been extended with a more detailed comparison for observations and modeling.
 - 11. Page 4, line 25. Figures 2 and 3 now are plotted in color.

- 12. Page 5, lines 10 and below. Figure 4 captures are corrected top panels are for model output, and low panels for MERRA data.
- 5 Thank you again for taking the time to review our manuscript.

With respect,

Yu.M.Timofeyev, S.P.Smyshlyaev, Ya.A.Virolainen, A.S.Garkusha, A.V.Polyakov, M.A. Motsakov, O.Kirner.

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Reply to reviewer 2

Dear Referee,

5 Thank you for your comments on the paper and constructive recommendations. We have tried to follow your suggestions and have utilized most of them. Following we mention how the manuscript has been changed according to your comments.

Major issue:

- 10 The conclusion about the role played by chemical and transport processes is not strongly supported by model results. It is mostly based on general knowledge of the different ozone time scales and not strongly supported by model results. The authors show that the RSHU model simulates some enhancement of PSC occurrence and ozone loss rate in the low TCO regions, but do not use it to support or reject the importance of chemical ozone loss. I think it makes sense to run specially designed model experiment (say the run without heterogeneous chemistry) to support purely dynamical nature of the low
- 15 TCO events or explain why such runs cannot be performed.

ACCEPTED

Two additional numerical experiments were carried out with RSHU CTM to confirm the conclusions

- 20 about the dominant role of the dynamical processes in the observed short-term ozone loss: one did not take into account the formation of polar stratospheric clouds in the Arctic zone, and the second did not take into account the chemical destruction of ozone to the north of the northern polar circle. A comparison of the three model experiments for the three stations considered in this paper is shown in Fig. 6. The results of model experiments have shown that the main features of the short-term ozone loss
- 25 are reproduced even without taking into account chemical destruction within the polar zone. At the same time, the influence of chemical processes becomes noticeable at the end of March, especially for Pechora.

Minor issues.

1. Page 1, Line 15: "unlikely" sounds too weak for the abstract.
2. Page 1, Lines 23-24: I would avoid using the same sentences in the introduction and conclusions. Potential readers could wonder if the minor role of heterogeneous chemistry is well known than why to tackle this issue again?

3. Page 1, Line 28: 2015/2016

4. Introduction: The motivation for the presented study should be stronger. The authors should emphasize the necessity to analyze new instruments and exploit two different models.

5. Ozone depletion: I understand this as chemical processes. However, low TCO events can be explained by the

5 transport of low ozone to the considered location from the area inside polar vortex, where the ozone is small because of suppressed influx from the ozone production area. The authors concluded that the contribution of chemical destruction is small. Maybe than the ozone depletion term is not perfectly correct?

6. PSC: The ozone depletion via heterogeneous chemistry strongly depends on the availability of liquid sulfate aerosols. How they are treated in the models? The authors show PSC area. Does it include all kind of PSCs or just NAT?

7. Section 3: The exploited models use different meteorological reanalysis, therefore the difference in the results can be attributed to either model or reanalysis features. Would it be possible to attribute more precisely the difference between model results?

8. Page 5, lines 11-13: I am not completely agree with this statement. Does it mean that low TCO inside polar

15 vortices will not take place without heterogeneous chemistry and chlorine activation. I think the role of transport is more important.

9. Page 5, last paragraph: In the present form it is not instructive. See my major comment.

10. Figures 4,5: The numbers are too small and hardly visible.

11. Figure 5: My impression is that the ozone loss about 10(8) molecules per second is too large. Please, check.

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ACCEPTED

1. Page 1, line 15: The sentence corrected with replacement "unlikely" to more strong statement. 25

2. Page 1, lines 23-24. The sentence modified with attention shifted from the statement to a question whether chemical destruction on the surface of polar stratospheric clouds, for which a long existence of PSCs is necessary, to be responsible for the observed anomalies, or other factors, especially dynamic ones, would have a greater effect on the observed features.

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3. Page 1, line 28. Corrected form 2016 to 2015/2016.

4. Introduction. The motivation extended with a justification of the need for additional measurements and numerical model experiments.

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5. The term "ozone depletion" almost everywhere in the paper is replaced by "ozone loss".

6. A short description of the PSC formation and evolution code with appropriate references is added to the section 3. The code accounts for STS, NAT and ICE particles formation on the base of sulfur40 aerosol.

7. The motivation for using two different models is added. An additional numerical experiment using the ERA-INTERIM reanalysis data was performed with the RSHU model in order to compare the effect of different meteorological data on the comparison of the results of numerical modeling and local observations. The results of a comparison of numerical modeling and observations, as well as comparisons between models with different meteorological data, are expanded.

8. Page 5, lines 11-13. The sentence has been modified with a shift to a chance of heterogeneous ozone destruction which is checked at the following discussion. "This is a result of dynamical isolation, which leads to stratospheric cooling and potentially may cause ozone depletion as a result of heterogeneous chemical reactions on PSCs particles leading to chlorine activation."

10 9. Page 5, last paragraph. Results of additional numerical experiments without PSC processing included are added into discussion to demonstrate the prevalent role of dynamical processes in the observed short-term ozone loss.

10. Figures 4 and 5 are corrected to make numbers more visible.

11. Figure 5. Corrected to 10(-8). Actually this means that ozone loss coefficient was multiplied by

15 10(8) before plotting.

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Thank you again for taking the time to review our manuscript.

With respect,

Yu.M.Timofeyev, S.P.Smyshlyaev, Ya.A.Virolainen, A.S.Garkusha, A.V.Polyakov, M.A. Motsakov, O.Kirner.

20 REVISED PAPER WITH CHANGES MARKED

Case study of ozone anomalies over northern Russia in the 2015/2016 winter: Measurements and numerical modeling

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Abstract. Episodes of extremely low ozone columns were observed over the territory of Russia in the Arctic winter of 2015/2016 and the beginning of spring 2016. We compare total ozone columns (TOC) obtained using different remote sensing techniques (satellite and ground-based observations) and results of numerical modeling over the territory of the Urals and Siberia for the above period. We demonstrate that the provided monitoring systems (including new Russian Fourier-spectrometer IKFS-2) and modern 3-dimensional models are able to capture the observed TOC anomalies. However, the results of observations and modelling show discrepancies of up to 20-30% in TOC measurements. Analysis of the role of chemical and dynamical processes demonstrates that it is unlikely that observed short-term TOC variability may be a result

10 of local photochemical destruction loss initiated by heterogeneous halogen activation on particles of polar stratospheric clouds that formed under low temperatures in the mid-winter.

1 Introduction

Abnormally low values of total ozone columns (TOC) were recorded in January-February 2016 in the polar region of the Northern Hemisphere (Zvyagintsev et al., 2016; Manney and Lawrence, 2016). Observed low values were recorded long

- 15 before the beginning of spring, when chemical destruction of ozone occurs periodically in the Northern Hemisphere as a result of a strong vortex and the long existence of polar stratospheric clouds (PSCs) (Manney et al., 2011). Early anomalies in TOC indicate that it is unlikely that they may be caused by chemical disruption after the heterogeneous activation of chlorine and bromine gases on the surfaces of PSCs particles during 2016 winter make one wonder whether chemical destruction on the surface of polar stratospheric clouds, for which a long existence of PSCs is necessary, to be responsible
- 20 for the observed anomalies, or other factors, especially dynamic ones, would have a greater effect on the observed features. The analysis of meteorological conditions during the 2015/2016 winter showed that during this period the lower polar stratosphere was extremely cold, which created a potential for a record ozone hole depletion in the spring of 2016, but a strong sudden stratospheric warming in early March 2016 destroyed the polar vortex and prevented formation of a spring ozone anomaly (Manney and Lawrence, 2016). Nevertheless, during the entire winter of 2016/2016 in the northern part
- 25 of Russia, the ozone content was lower than in previous years, and the depth of short-term ozone anomalies in January and February 2016 was comparable to the depth of the ozone mini-holes of the spring 2011.

Over recent decades, investigation of total ozone time-scale variations demonstrated regular occurrence of the spring deep ozone depletion over the Antarctic region. This phenomenon was called the "ozone hole". In the Northern Hemisphere, similar to the southern hemisphere polar column ozone depletion loss has been observed on smaller scale as well as over

30 shorter time intervals, for example in 2011 (Manney et al., 2011; Balis, 2011). For episodes with extremely low TOCs (less than 220 Dobson units) these phenomena were called "ozone mini-holes" (Millan and Manney, 2017). Observation and

prediction of the occurrence of episodes with abnormally low ozone content close to "mini-holes" is both crucial for the investigation of its nature and for the prediction of potential increase of UV-radiation on the Earth's surface. Unusually sharp and repetitive TOC depletion loss was observed over the territory of the Urals and Siberia in the first quarter of 2016. In some cases, the TOC depletion loss reached 40-50% in comparison with climatic values (Zvyagintsev et al., 2016).

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In this paper, we study the episodes of low TOCs over some Russian stations in January and February 2016 based on remote sensing observations and results of numerical modeling.

2 Total ozone column measurements over Russia during winter 2016

Monitoring of the total ozone level is provided by various ground-based remote sensing systems (Brewer and Dobson spectrophotometers, M-124 filter ozonometers, DOAS, Microwave and IR methods, lidar measurements) and by various satellite systems (https://disc.gsfc.nasa.gov/; Timofeyev and Vasiliev, 2008; Staehelin et al., 2001). According to regular extensive validation programs (Balis et al., 2007; Boynard et al. 2016; Garkusha et al., 2017), total ozone measurement errors can be from 1–2 to 10% depending on the method, device, time and place of the measurements.

We analyzed the total ozone data of the first quarter of 2016, obtained by the basic Russian ground-based ozonometer M-124 and satellite instruments OMI and SBUV (recording outgoing solar reflected and scattered spectra of UV radiation),

- 15 IASI and a new Russian instrument IKFS-2, recording outgoing atmospheric thermal IR radiation. The features of such satellite instruments as OMI, SBUV and IASI and the Russian ground-based ozonometer M-124 are well-known (Balis et al. 2007; Bhartia et al., 2013; Kroon et al., 2008; Viatte et al., 2011; Boynard et al., 2016). Independent assessments of TOC measurement errors (Virolainen et al., 2017) showed values of 3.3–4.1 % for IASI, 2.0–3.5 % for M-124, and 1.9–2.1 % for OMI instruments. The infrared Fourier-transform spectrometer IKFS-2 on-board the satellite "Meteor-M" was launched in
- 20 July 2014. IKFS-2 was preeminently designed for temperature-humidity sounding of the atmosphere and for measurement of some climatically important gases, including ozone. Detailed description of characteristics of IKFS-2 is given by Golovin et al. (2014). The advantage of the IKFS-2 and IASI instruments is its ability to conduct measurements in the absence of sunlight, which is especially important for polar regions, where the polar night exists for a long time, during which the work of solar radiation measurement devices is impossible.
- The description of the IKFS-2 measurement interpretation methodology, as well as estimates of errors in measurements of TOCs for cloudless and cloudy atmosphere, are given in the works (Garkusha et al., 2017; Garkusha et al., 2018). The technique of interpretation, based on the method of artificial neural networks (ANN), is described in detail in the paper (Garkusha et al., 2017). The approximation of the solving operator of the inverse problem by a three-layer perceptron is used, the activation function of the neurons of the hidden layer is the hyperbolic tangent, the output neuron is linear. The
- 30 main feature of the technique is the use as predictors of principle components (PC) the spectra measured by IKFS-2. The set of predictors consists of 25 PC of the entire measured spectrum (660-2000 cm⁻¹), 50 PC only of the ozone absorption band and the measurement zenith angle. For ANN training, the results of TOC measurements using the OMI instrument from the AURA satellite were used (McPeters et al, 2015). Estimates of the error in determining the TOCs with IKFS-2 are on the

average in the range 2-6%. The largest differences (up to 10%) are observed in the southern polar latitudes in the presence of an ozone hole over Antarctica.

In the first quarter of 2016, three long short-term periods of essential deviations of the average daily TOCs from average long term values with significantly lower daily TOCs compared to the climatologically average values for the period from

- 5 1979 to 2017, were registered over the territory of Russia. TOCs decreases reached: 39–52% (in 26.01–01.02 over the Northern regions of the Urals and Siberia), 30–50% (in 20.02–03.03 over Northern Siberia), 27-39% (in 09–19.03 over Central Siberia) of average values (191–257, 227–321, 257–321 DU, for these three periods, respectively) (Zvyagintsev et al., 2016). Extremely low winter TOC values (episodically less than mini-hole threshold) were observed over the northern regions of the Urals and Siberia for the first time. During January 27–31 TOCs smaller than 220 DU were recorded at
- 10 Russian ozonometric network stations using M-124 measurements (Pechora, 65°N, 57°E; Khanty-Mansiysk, 61°N, 69°E; Turukhansk, 66°N, 88°E; Round, 64°N, 100°E) and by OMI devices on the board of Aura satellite.

Figure 1 taken from (Garkusha et al 2018) depicts the spatial distribution of TOCs in February 23-27, 2016, based on measurements of two instruments of the same type - IKFS-2 and IASI. The figure shows good agreement between two independent satellite measurements.

Figure 2 presents the evolution of TOCs measured at three ground-based observational stations: Tura (61° N, 100° E), Pechora, and Khanty-Mansyisk. The comparison allows us to draw the following conclusions:

(a) All instruments and measurement methods generally provide a good description of the main features of TOC time variations, including observed ozone depletion-loss. For the whole period of comparison, the average differences in the results obtained by different types of measurements in most cases are 1-5%, with standard deviations of 3-8%.

(b) The only exception is the IASI measurements in Khanty-Mansiysk and Tura, for which the standard deviations of the differences with ground-based measurements in the first quarter of 2016 reach 12%. In addition, at the Tura station, IASI data overstates the M-124 measurements by 11% on average.

(c) All satellite data overestimate the values of TOC in comparison with ground-based measurements during the short-term periods of ozone depletion loss. This is, possibly, due to the fact that the optimal retrieved solution is constructed not only from atmospheric radiation spectra that have been measured, but it also employs *a priori* information about TOCs. This then described the mean state of ozone concentrations and not a mini hole event. This *a priori* information does not account for particular ozone profile and may cause distortions in the estimation of the local TOC. To exclude this effect, it is necessary to improve the *a priori* information by incorporating the ozone total column amount with the appearance of strong ozone anomalies-making it dependent on the type of ozone profile, characteristic for the season and the region and observations.

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3 Comparison of total ozone column measurements and numerical modeling

Also relevant to this issue is the comparison of observational data with the results of numerical modeling. Values of TOC were calculated by two three-dimensional atmospheric chemistry models, which take into account observed variations of meteorological parameters based on re-analysis of the measurement results, the chemistry-climate model (CCM) EMAC

- 5 (ECHAM/MESSy Atmospheric Chemistry model) (Jöckel et al., 2006) and the Russian State Hydrometeorological university chemistry-transport model (RSHU CTM) (Smyshlyaev et al., 2017). The motivation for using two different models is to try to assess the impact of the interactive coupling between physical and chemical processes available in the CCM relatively offline meteorological data using in the CTM. In addition, the models have different spatial resolution, which makes it possible to estimate the effect of model resolution on the comparison with the observations related to the
- 10 local points.

The EMAC model is a numerical chemistry and climate simulation system that includes tropospheric and middle atmosphere processes (Jöckel et al., 2010). It uses the second version of the Modular Earth Submodel System (MESSy2). The core atmospheric model is the 5th generation European Centre Hamburg general circulation model (ECHAM5, Roeckner et al. 2006). The core model, ECHAM5, uses a spectral transform technique, the so-called T-value indicating the degree of

- 15 triangular spectral truncation. For the present study, we applied EMAC (ECHAM5 version 5.3.02, MESSy version 2.52) in T42 resolution; i.e., with a spherical truncation of T42 (corresponding to a quadratic grid of 2.8 x 2.8 degrees, respectively, in latitude and longitude). Vertically, the model resolves the troposphere, stratosphere and lower mesosphere (39 hybrid levels from the surface up to 0.01 hPa, about 80 km). We applied a Newtonian relaxation technique (Nudging) to our model simulation with the help of the ERA-INTERIM reanalysis data set (Dee et al., 2011) to improve consistence between the
- 20 simulated and observed temperature and wind fields responsible for the dynamical impact on ozone distribution. A detailed description of the EMAC model and its applications can be found in (Righi et al., 2015, Virolainen et al., 2016).

The global RSHU CTM is based on the Institute of Numerical Mathematics and Russian State Hydrometeorological University (INM RAS – RSHU) CCM (Galin et al., 2007), but meteorological fields are not calculated but specified from the ERA-INTERIM or Modern-Era Retrospective Analysis for Research and Applications (MERRA) reanalysis (Rienecker et

- 25 al., 2011) reanalyzes. The use of different reanalysis data made it possible to compare their effect on the observed shortperiod variability of the ozone content at the observation stations under study. The model The RSHU CTM has 5 x 4 degrees horizontal resolution in longitude by latitude and 31 vertical sigma levels from the surface up to approximately 60 km. The distribution of the oxygen, hydrogen, nitrogen, chlorine, bromine and carbon gases are calculated in the manner described by Smyshlyaev et al. (1998). PSCs formation and evolution is taken into account according to Smyshlyaev et al. (2010).
- 30

- Both models sufficiently describe time variations of the total ozone content. On average, the RSHU model provides 1–2% smaller values of the TOC than those observed by OMI. EMAC, conversely, exceeds the OMI measurements by 7–9%.

The analysis of comparison between modeling and experimental (OMI, version TOMS) leads to the following conclusions (Fig.3):

The standard deviations for both models are 6–7%. This approaches the standard deviations between different types of measurements of the total ozone content during the examined period.

- EMAC better describes the TOC variations during some depletion periods than the RSHU CTM model: at the Khanty-Mansiysk station standard deviation stood at 4–5% for EMAC model whereas the RSHU model ranged between 6 and 8%.

5 At the Tura station during the January minima, on the contrary, the RSHU model is in better agreement with OMI measurements (3% vs. 7%). Neither model describes the observed January mini-holes at the Pechora station (standard deviations reach 12–15%).

- On certain days, the differences between measurements and modeling can be up to 20–30%. Models often overestimate the total ozone content measured by the OMI instrument (especially the EMAC model).

- In general, the comparison of calculations with two different models which use ERA-INTERIM reanalysis by different manner, demonstrated that both the interactive coupling between physical and chemical processes and higher spatial resolution do not have a principal influence on the quality of reproduction the short-term column ozone variability at local points. Both models demonstrated not bad qualitative correspondence to the OMI satellite observations, while for some local points and time periods the best correspondence was shown by the EMAC CCM, and for others by RSHU CTM.
 Comparison of the results of the calculations with the RSHU CNVI with various re-analysis data showed that for MERRA
- 15 Comparison of the results of the calculations with the RSHU CNVI with various re-analysis data showed that for MERRA data, the column ozone is systematically lower than when using EPA-INTERIM data.

4. Analysis of the processes that define observed ozone variability over Russia during the Arctic winter of 2015/2016

The role of chemical and dynamic processes in the observed TOC variability over Russia was assessed based on the 20 RSHU CTM calculations. Two days with the lowermost TOCs registered at all stations were selected for extended analysis. These days are January 27, 2016 (day 27) and February 19, 2016 (day 50) (Fig.3). Results of RSHU CTM simulations for these days are presented in Fig.4 for column ozone (top figures) together with the MERRA temperature data averaged for the lower stratosphere (14–25 km) (bottom figures). The regions with low TOCs are consistent with the low stratospheric temperatures. This is a result of dynamical isolation, which leads to stratospheric cooling and potentially may cause ozone 25 depletion as a result of heterogeneous chemical reactions on PSCs particles leading to chlorine activation.

The surface area of the PSCs for the days with low stratospheric temperature and low column ozone episodes are presented in Fig. 5 (top panel). Enhanced PSCs surface area is located at the same regions where low stratospheric temperatures were registered. This is obvious consequence of stratospheric cooling and may lead to heterogeneous chlorine and bromine activation followed by ozone depletion similar to the Antarctic ozone hole formation (Solomon, 1999). In order

30 to evaluate local ozone destruction significance for the observed TOC depletion loss, the photochemical ozone loss coefficient (s⁻¹) (Jacobson, 2005) (rate of ozone loss divided by the ozone concentration: $\Lambda_{O3} = L_{O3} / N_{O3}$, where L_{O3} - is

photochemical ozone loss (mol/s/cm³) and N_{O3} is ozone concentration (mol/cm³)), calculated with the RSHU CTM, is presented in the bottom panel of Fig.5.

The location of zones with enhanced ozone destruction is close to the regions with estimated low TOCs, but is not fully consistent. In addition, the minimum local photochemical ozone lifetime, estimated as a reciprocal of the ozone destruction

- 5 coefficient, is about 200 days under these days' conditions. Such a long photochemical lifetime of ozone may be treated as a sign of the unlikeliness that the observed short-term ozone variability may be a result of local photochemical destruction initiated by heterogeneous chlorine and bromine activation on the particles of PSCs that formed in these regions. On the other hand, simultaneous low stratospheric cooling and low column ozone at the same locations may be caused by dynamic divergence that leads to heat and mass deficit, similar to polar vortex isolation (Solomon, 1999). Another confirmation of the
- 10 prevalent dynamical nature of the observed episodes with low ozone concentration is their formation during polar night recorded during December 2015 and first part of January 2016 when photochemical destruction is negligible.

In order to check the conclusion about the dominant role of the dynamical processes in the observed short-term ozone loss two additional numerical experiments were carried out with RSHU CTM : one did not take into account the formation of polar stratospheric clouds in the Arctic zone, and the second did not take into account the chemical destruction of ozone to

15 the north of the northern polar circle. A comparison of the three model experiments for the three stations considered in this paper is shown in Fig. 6. The results of model experiments have shown that the main features of the short-term ozone loss are reproduced even without taking into account chemical destruction within the polar zone (red curve). At the same time, difference between results of the model experiments with and without polar chemical ozone destruction (claret curve) depicts that the influence of chemical processes becomes noticeable at the end of February, especially for Pechora station.

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5. Summary

Data analysis and numerical model experiments have been used to analyze the low TOCs recorded over Russia during the 2015/2016 winter. Ozone anomalies were observed over the territory of the Urals and Siberia in winter and the beginning of spring 2016. In this paper, we compare TOCs obtained using different measuring methods (satellite and ground-based observations) and results of numerical modeling (for the stations Khanty-Mansiysk, Tura, and Pechora) for the above period. It is shown that existed monitoring systems (including Russian Fourier- spectrometer IKFS-2) and modern 3-dimensional models provide a good description of the occurrence of the TOC anomalies. However, results of observations and modeling diverge on particular days by as much as 20-30%. Analysis of the role of chemical and dynamical processes in the observed ozone variability over the Russian Federation was based on the RSHU CTM calculations. This analysis demonstrated that it

30 is unlikely that local photochemical destruction initiated by heterogeneous halogen activation on the particles of PSCs that formed under registered low temperatures may be responsible for short-term local ozone destruction. The prevalent reason for the observed low TOCs may be dynamical flux divergence out of regions with observed low ozone content (Smyshlyaev et al., 2017).

Acknowledgments

The comparison and analysis of different experimental and simulated total ozone column data was supported by the Sussian Science Foundation (Grant #14-17-00096). The modeling of Arctic processes with the RSHU CTM was supported by the Russian Foundation for Basic Research (Grant #17-05-01277). The interpretation of Russian Fourier- spectrometer IKFS-2 spectra was supported by the Russian Foundation for Basic Research (Grant #17-05-01277). The interpretation of Russian Fourier- spectrometer IKFS-2 spectra was supported by the Russian Foundation for Basic Research (Grant #17-05-00768). The discussions of the results and the retrieval of EMAC model data was supported by Saint Petersburg University (Grant #11.42.690.2017). The authors thank A. M. Zvyagintsev from the Central Aerological Observatory (Dolgoprudny, Russia) for providing the ozone

10 measurements from the ground-based network. Both the simulation data and measurement results necessary to reproduce the comparison are available from the authors upon request (<u>vana.virolainen@spbu.ru</u>).

References

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Figure 1. Spatial distributions of the total ozone columns in February 23 - 27, 2016, based on measurements of two instruments of the same type - IKFS-2 (left) and IASI (right). (Garkusha et al, 2018).



Figure 2: Total ozone measurements provided by the OMI, M-124, IKFS-2, IASI, and SBUV for Khanty-Mansiysk, Tura, and Pechora stations.



Figure 3: Total ozone measurements provided by OMI and modeling results from EMAC and RSHU for the stations Khanty-Mansiysk, Tura, and Pechora.



Figure 4: Column ozone (Dobson units) for days with minimum local registered values, simulated with the RSHU model (*top panels*) and temperature of the lower stratosphere (K) from MERRA reanalysis for the same days (*bottom panels*).



Figure 5: Calculated with RSHU CTM low stratospheric polar stratospheric clouds surface area $(10^8 \text{ cm}^2/\text{cm}^3)$ for days with minimum local registered column ozone values (*top panel*) and averaged for the low stratosphere ozone loss coefficient (10^{8}s^{-1}) for the same days (*bottom panel*).



Figure 6: Column ozone variability modeled with RSHU CTM for different scenarios for the stations Khanty-Mansiysk, Tura, and Pechora and difference between scenarios with no polar chemistry and full PSC processing included