Dear Editor, dear 3 Reviewers,

We answered all questions and provide all our earlier individual answers in this document. In the new version of the manuscript the changes due to the remarks of reviewer one are in red color, those to reviewer 2 in green and those to reviewer 3 in blue and hope that this provides a quick and easy overview.

We thank all three reviewer for the careful reading and the generally positive reviews. Our detailed answers (as provided individually to all three of you) are:

Answers to reviewer 1: (red sentences in new version)

Quote line 65:

The warming of Spitsbergen and the possible relation to changed atmospheric circulation pattern was shown here: (Isaksen et al. 2016)

https://agupubs.onlinelibrary.wiley.com/doi/full/10.1002/2016JD025606

The impact of the West Spitsbergen current on the local climate is described here

https://rmets.onlinelibrary.wiley.com/doi/full/10.1002/joc.2338

We include this quote in the new version of the manuscript

Isaksen, K.; Nordli, O.; Forland, E.J.; Lupikasza, E.; Eastwood, S. and Niedzwiedz, T. Recent warming on Spitsbergen—Influence of atmospheric circulation and sea ice cover, J. Geophys. Res. Atmos., 121, 11,913–11,931, doi: 10.1002/2016JD025606. 2016

Walczowski, W.; Piechura, J. Influence of the West Spitsbergen Current on the local climate. nt. J. Climatol. 31: 1088–1093, doi: 10.1002/joc.2338. (2011)

Quote line 125:

The possibility to derive differences on aerosol properties comparing neighboring stations for Spitsbergen is described in the coming paragraph with corresponding quote. For this reason we include a more general quote here (Sakerin et al. 2010)

https://link.springer.com/article/10.1134/S1024856010020028

The importance of Arctic intercomparison campaigns with error estimation was provided by Mazzola et al. 2011

https://repositorio.aemet.es/bitstream/20.500.11765/11692/1/Mazzola-Atm-Env-2011.pdf

We include this quote in the new version of the manuscript

Sakerin S.M., Kabanov D.M., Nasrtdinov I.M., Turchinovich S.A., and Turchinovich Yu.S. The results of two-point experiments on the estimation of the urban anthropogenic effect on the characteristics of atmospheric transparency // Atmospheric and Oceanic Optics, 2010, Vol. 23, No. 2, p. 88–94. DOI: 10.1134/S1024856010020028.

Mazzola, M.; Stone, R.S.; Herber, A.; Tomasi, C.; Lupi, A.; Vitale, V.; Lanconelli, C., Toledano, C.; Cachorro, V.E.; O'Neill, N.T.; Shiobare, M.; Aaltonen, V. et al. Evaluation of sun photometer capabilities for retrievals of aerosol optical depth at high latitudes: The POLAR-AOD intercomparison campaigns. Atmosph. Environm. doi: 10.1016/j.atmosenv.2011.07.042. 2011

Paragraph lines 135 – 140: on the differences in AOD over both stations.

In his works [Toledano et al., 2012; Pakszys and Zielinski, 2017] had already compared AOD, measured at the neighboring stations Hornsund and Ny-Ålesund. The difference in the seasonally and annually average values of AOD between these stations reached 0.01-0.02, mainly due to situations with high atmospheric turbidities. We obtained about the same result after hourly average AOD in Ny-Ålesund and Barentsburg were compared (when measurements were within one hour of each other).

How can the difference in AOD between the neighboring regions be explained?

Even without local anthropogenic impact, the difference in AOD between stations, separated by mountains and distances 100 km (or longer) apart may be due to the Arctic Haze phenomena and transports of smoke plumes. (They are not conservative and homogeneous structures, blown by wind without changes). The Arctic Haze or plumes may be observed in the region of any station, and may be observed (to a lesser extent) or not (due to spatial inhomogeneities) at another one. When AOD, measured at two stations within an hour (as in our work), are compared, the number of such inhomogeneous situations can only be reduced partly sometimes, but can never be eliminated at all.

Resort to data from trajectory analysis or to any correction (or shift) in time makes no sense for two reasons. First, this cannot be made due to spatial inhomogeneities in AOD: for instance, there was the thickest part of the plume in the region of one station, and a thinner part in the region of another one. Second, the AOD observations are not continuous, being carried out only in situations when Sun is not covered by clouds. That is, the measurements were in the period of AOD maximum at one station, and only during AOD decay or commencing AOD growth at another one.

Taking into consideration the Reviewer's comment, we corrected slightly the text in this paragraph:

"Comparison of measurements with the two photometers showed a large dispersion of the data under the conditions of strong atmospheric turbidities, namely, during outflow of smoke plumes from forest fires and in the Arctic Haze situations. Due to large spatial inhomogeneity of these structures, AOD, measured in two regions, may strongly differ, making the comparison incorrect".

Line 165: thanks, we added a short explanation to the Ångström formula in the new manuscript:

The attenuation of radiation by atmospheric aerosol varies as a function of wavelength, depending on sizes and refractive index of aerosol particles. To characterize the AOD, measured at different wavelengths, the Ångström formula is widely used:

 $\tau^a(\lambda) = \beta \cdot \lambda^{-\alpha}$

(1)

where β and α are the approximation parameters of the spectral dependence of AOD; β is the turbidity coefficient, which is close in value to AOD at the wavelength of 1 μ m; and α is the selectivity exponent (power-law decay).

Line 180:

Thanks – we explain now that m corresponds to β and n to α in eq. 1.

Line 185:

Thanks we changed the wording: with independent data we mean additional information. We write:

"But, precisely what caused changes in the selectivity of AOD is almost impossible to determine without the use of additional information like e.g. aerosol in-situ measurements".

Line 300:

In this paragraph, we compare two periods of measurements at a **<u>single</u>** station (*Ny-Ålesund*) and indicate a tendency toward a small AOD decrease in 2011-2018 relative to 2002-2010. The location of this station, orography, or something else did not change.

By the wording "no explicit predominance" we wanted to express that no single value for the variation coefficient for fine or coarse mode for the three places and times: Ny-Ålesund: early, Ny-Ålesund: later, Barentsburg: later dominate over the others. This means that there is no clear shift in aerosol properties neither in time nor from Barentsburg to Ny-Ålesund.

We rewrite the sentence for clarity:

No explicit predominance of the variation coefficients for any AOD component can be seen.

The relative variations $\tau^{f}_{0.5}$ and τ^{c} are about the same: their variation coefficients V are 14-29%. Neither AOD component shows a clear predominance of variation coefficients.

We fixed the Fig. 1 and 9, as suggested by the Reviewer.

Answers to reviewer 2: (green sentences in new version)

L 14: we do not use a symbol in the abstract any longer

L 101: thanks – corrected

L 131 "Larger AOD values occur"

Yes, this difference in AOD is between two stations, not between seasonal or annual values, but between specific situations. For a more clarity, we corrected somewhat the sentence:

"Large differences in AOD between these stations occur in periods of the Arctic haze and outflows of smokes from forest fires".

L 143: thanks – corrected!

L 142-145: decreasing AOD towards IR and variability

This is a good remark. However, we considered this: In Fig 11 it will be shown later that the AOD at 0.38 μ m is typically less than a factor of 4 larger than the AOD at 0.87 μ m, while the difference between the sites is almost a factor of 5.

We add (new part in bold)

This feature **is real despite the decreasing AOD at longer wavelengths and** indicates that fine aerosol is more abundant in the atmosphere of Barentsburg

L 206: Thanks we use always EM in the new version.

L 211: thanks - corrected

L215 – 216:

We clarify that the average difference in τ^c , calculated by different methods (and for different conditions), relative to the empirical method is, indeed, no higher than 0.007. While the standard deviations of the regression between the two compared τ^c values are in the range of 0.006-0.024 (see [Kabanov et al., 2016] for more detail).

L 261: disperse composition

By disperse composition we mean the size distribution and changed the wording in the manuscript.

L 265 – 269:

Here, there are two questions.

1. Yes, in this case we used IM1, which has much better characteristics of the interrelation with EM data. You are right: IM1 also depends on the choice of the refractive index. But this is not important because at the last stage of implementation of this method (see Line 239-240), we also use the regression relation and select the approximation parameters. That is, we could specify a slightly different refractive index and select slightly different approximation parameters for the linear regression. We made so and obtained about the same result. In principle, simpler methods of the τ^c calculation could be used (e.g., RM2). From Table 2 it can be seen that the errors of different methods differ insignificantly.

2. Of course, seasonal and interannual variations are easier to analyze in the optical characteristics: they are just two, τ^c and τ^f . Analysis of variations in microphysical composition of aerosol is a more complex problem: it will be necessary to consider the particle distribution functions (i.e., changes in two or three parameters for each aerosol fraction) and, moreover, the refractive index, which is at all unknown, in this case.

L 303:

We corrected somewhat two sentences in this paragraph:

"The relative variations in $\tau_{0.5}^{f}$ and τ^{c} are about the same: their variation coefficients V are 14-29%. Neither AOD component shows a clear predominance of variation coefficients".

Table 3: line deleted, thanks!

L 335 – 337:

The seasonal and interannual AOD variations were analyzed individually over a full dataset in each region. That is, no selection of data with identical hours of measurements was performed in this case.

L 359:

To identify smoke outflows (in the cases of large AOD values), we used data on back trajectories of air mass motion (HYSPLIT) in combination with satellite maps of fire centers (temperature anomalies).

Table 4: thanks - corrected

L 373:

We clarify that the average and modal (most probable) values are different statistical characteristics. The first sentence of this paragraph is about the average values, presented in Table 4 and in Fig. 10. In turn, the second sentence is about modal values, which are presented in Fig. 12a. Thus, there is no contradiction in that the average values decrease by the amount 0.015-0.016, while modal values decrease from 0.07 to 0.03.

Figure Caption 3: thanks – corrected

Figure 9:

The solid line is the unconstrained fit (Y=aX+b). The dotted line is the fit through the origin (Y*aX). We explain this in the new version.

Answers to reviewer 3: (blue sentences in new version)

Line 27.

In the phrase about 10(20) years there is no need in making a reference: it is just a reminder about the well-known fundamentals of statistical data analysis. To identify the regularities of variations in climate characteristics, which depend upon many processes on various variability scales, the length of observation time series should be many times (an order of magnitude or more) larger than the periodicity sought. For instance, to determine the character of the seasonal behavior, the length of observation time series should be of the order of 10 years. This will be shown later in our section 3.1, figure 6. However, under the conditions of reforming climate system, the statistical estimates may give a distorted or incomplete understanding of the studied process because of the new variability factors emerged. Taking into consideration your comment, we modified one sentence (Line 28-29) a little:

"However, under the conditions of changing climate system, a time series 10 years (or even 20 years) long may turn out to be insufficient to identify correctly the tendencies or periodicities in variations of aerosol characteristics".

Line 49.

You are probably right. The impact of the Kasatochi eruption on sun-photometer data only appeared short as the season of observations ended early Oct. We rewrote the sentence:

new

The effects of less powerful volcanic eruptions on the Arctic atmosphere are short-term to mid-term (some weeks in duration). For instance, AOD increase on Spitsbergen was observed after the eruptions of volcanoes Kasatochi (August / Sep 2008) and Sarychev (after July 2009) [Hoffmann et al., 2010; Toledano et al., 2012].

old

The effects of less powerful volcanic eruptions on the Arctic atmosphere are short-term (a few days in duration) and comparable to those due to smokes from forest fires. For instance, AOD increase on Spitsbergen was observed after the eruptions of volcanoes Kasatochi (August 2008) and Sarychev (July 2009) [Hoffmann et al., 2010; Toledano et al., 2012].

Line 142.

(a) This phrase does not refer directly to Fig. 1, because it is in the next paragraph. Difference in the data from quasi-synchronous AOD measurements between two stations was already considered in our previous work [Kabanov et al., 2018] (see reference citation in Line 134), so only a conclusion is presented here. For more clarity, we gave the reference citation again in this sentence:

"A comparison of the statistical characteristics showed that the average AOD values are a little larger in Barentsburg than in Ny-Ålesund [Kabanov et al., 2018]".

(b) We think that it is a bad idea to display the average difference in the data between two stations in Fig. 1. But, if the Referee considers that this is mandatory, we prepared the following variant of the figure:



Fig. 1. Scatter diagram of AOD measurements in two regions using SP1A (Ny-Ålesund) and SPM (Barentsburg) photometers. Solid line: unconstrained linear regression; dotted line: regression through origin (0,0); green indicates the average value of the data difference and the standard deviation

<u>Line 145</u>. Taking into consideration your comment, we modified one sentence (the word **error** was replaced by **uncertainty** and links were added):

«At the same time, we note that the AOD differences are minor (comparable with uncertainty of determining AOD – about 0.01-0.02 [Kabanov et al. 2009; Sakerin et al., 2013]), and the interdiurnal AOD variations in the two regions are coordinated in character (correlation coefficients are 0.83-0.89)».

Line 148.

(a) Yes - the Δ symbol indicates the average difference between the measurement data of two photometers (or AOD at two stations) at different wavelengths. This is not a measurement error, but the difference in physical characteristics in the two regions. The error (or rather, uncertainty) of the AOD measurements is 0.01-0.02 (see Line 145).

(b) Table 2 shows statistical characteristics on a completely different issue: standard deviations σ and correlation coefficients R between τ^c values calculated by different methods.

Line 168. Although Angstrom's formula is well known, we have added links at the request of the Referee:

«Numerous studies in different regions and atmospheric conditions showed that the formula (1) does describe well the wavelength dependence $\tau^a(\lambda)$ in the main range (0.34 – 1 µm) of AOD measurements [Angstrom, 1964; Shifrin, 1995; Eck et al., 1999; Cachorro et al., 2000; Schuster et al., 2006].

- 1. Angstrom A. Parameters of atmospheric turbidity // Tellus XVI. 1964, N 1, p. 64-75.
- 2. Shifrin K.S. Simple relationships for the Angstrom parameter of disperse systems // Appl. Opt., 1995, Vol. 34, 4480-4485.
- Eck, T.F., Holben B.N., Reid J.S., Dubovik O., Smirnov A., O'Neill N.T., Slutsker I., and Kinne S. Wavelength dependence of the optical biomass burning, urban, and desert dust aerosol // J. Geophys. Res., 1999, Vol. 104, 31333-31350.
- 4. Cachorro V.E., Duran P., Vergaz R. and de Frutos A.M. Measurements of the atmospheric turbidity of the north-centre continental area in Spain: spectral aerosol optical depth and Angstrom turbidity parameters // J. Aerosol Sci. 2000, Vol. 31, No. 6, pp. 687-702.

5. Schuster, G.L., Dubovik O., and Holben B.N. Angstrom exponent and bimodal aerosol size distributions // J. Geophys. Res., 2006, Vol. 111, D07207. doi:10.1029/2005JD006328.

Line 179.

The main point in this section was to show that the Ångström parameters are inconvenient for analysis of the causes for AOD variations. In particular, the parameter α depends on the relationship between two AOD components (t and t). Therefore, based on the parameter α , it is difficult to judge what (whether t or t) was the cause for the AOD variations. The dependence of α on the components t and t is more complex and explicitly nonlinear in character. A variant of this dependence in the form $\alpha = \ln[(\tau_{0.5}^f/\tau^c) + 1]$ is presented in Fig. 2. Seemingly, a better approximation of this dependence can be selected; for instance, "a second order behavior of alpha" can be used, as was done in the SDA method. However, this requires a separate study, and was beyond the scope of this paper. In this case, it was sufficient to show that α does depend on the relationship (t/τ), and we did so (see regression in Fig. 2).

Line 191. An explanation of the symbol P (confidence level) was added to this sentence: «The correlation coefficient between α and $\ln[(\tau_{\lambda}^{f}/\tau^{c}) + 1]$ is statistically significant and equal to 0.68 (confidence level P < 0.0001)».

"Confidence level P" is a standard and well-known statistical characteristic. Therefore, there is no need in providing a reference to any text-book.

Line 199. In this sentence, we have added refinement (in troposphere): «The lifetime of fine aerosol in troposphere is a few days; therefore, it can be transported long distances (hundreds and thousands of kilometers) away».

Line 214. You are right: the notation $\tau_{0.5}^{f}(t)$ may be unclear. Therefore, in this sentence and in the text below we made the following correction: $\tau_{0.5}^{f}(or t)$. We clarify that, within different methods, the researchers calculate anyway a single component (either $\tau_{0.5}^{f}$ or t^{c}), while the other is found as a residual of the total AOD (see Line 207-208).

Line 217. Thank you for your comment. We fixed the link (correct – [Kabanov et al., 2019a): «For the conditions of Arctic region (Spitsbergen), we performed an additional study [Kabanov et al., 2019a], concerning the selection of an optimal method of $\tau_{0.5}^{f}$ (or τ^{c}) estimation».

Line 221. According to the Referee's comment, we added two clarifying sentences (see paragraph above):

"... In the first regression method (RM1), τ^c is estimated using its interrelation with the parameter β (see formula (3) below). In the second method (RM2), the regression dependence of $\tau_{0.5}^f$ on the parameters α and β (see formula (4)) is used. Comparison of different methods ..."

Line 228: Thanks, we shifted the Table 2 and corrected the wrong layout.

Line 230. We added the link [Kabanov and Sakerin, 2016], as suggested by the Referee: «The disadvantage of the regression methods is that they require a preliminary data accumulation under the conditions of a specific region for determining optimal regression coefficients in equations (3) and (4) [Kabanov and Sakerin, 2016]».

Line 238.

It is not quite clear which part of the IM1 method is requested by the Referee to be described in a more detail. We will try to do so, but before we will give three clarifications. 1. In implementing IM1 (or IM2) it is entirely immaterial which (Sviridenkov's or another)

The implementing IMT (or IM2) it is entirely immaterial which (Sviridenkov's or another) method of solving the inverse problems is to be used. A large number of methods have been developed in the last half-century to retrieve the particle distribution functions from spectral measurements of AOD or aerosol extinction coefficients. That is, any accessible and verified algorithm of solving the inverse problem, making it possible to determine the distribution function (dV/dr), or (dS/dr), will be appropriate.

2. It is immaterial what (whether dV/dr or dS/dr) should be retrieved, because an interrelation with the optical characteristic, τ^f or τ^c , can be found for any of them. In addition, (dV/dr) and (dS/dr) are interrelated mathematically (either can be calculated if the other is known). Therefore, only one characteristic, (dV/dr), will be used below.

3. In this work, we used the (available to us) Sviridenkov's algorithm of solving the inverse problem, which is a modification of the Twitty algorithm. The algorithm of the (now died) Sviridenkov M.A. has been tested and successfully used for as long as 20 years by a few scientific groups from Russia. Because you encountered problems with the reference to the work of Sviridenkov, we asked our Editorial office to make this issue (No. 12, 2001) of the Journal publicly available. We note that the results, obtained using the Sviridenkov's algorithm (as a part of the IM1 and IM2 methods), well agree with the data from other techniques (EM, RM1, RM2, and SDA). Therefore, we have no grounds to not trust the Sviridenkov's algorithm.

Using the IM1 method as an example, we will explain the consecutive steps of its implementation.

<u>Ist step</u>. Based on any known method of solving the inverse problem (for a specified refractive index, type of the particle distribution function, and grid of radius ranges), the spectral AOD values are used to calculate the particle distribution function (dV/dr) or (dS/dr). <u>2nd step</u>. In the distribution (dV/dr) thus obtained we select its part referring to the fine (submicron) fraction, and, for it, calculate the total particle volume (V^{f}) through integration. The size (radius) integration limits for particles in the fine fraction are: left boundary is specified depending on specific features of the chosen inversion algorithm (usually $r \approx$

 $0.1 \,\mu$ m); and $0.4 \text{ or } 0.5 \,\mu$ m is taken as the right boundary.

<u>3rd step</u>. We consider the regression interrelation between particle volumes in the fine fraction (V^f) and the t^f values, calculated by the empirical method (EM). The interrelation thus obtained (Fig. 4a) is used to select the parameters of a linear regression equation which makes it possible to calculate the component t^f according to the particle volumes V^f . (Figure 4b illustrates the comparison of t^f values, obtained by the two (EM and IM1) methods).

Taking into consideration the Referee's comments, we provided the text about the IM1 method in a little more detail (two insertions):

(1) "This method is implemented in the following steps.

Ist step. Based on any known method of solving the inverse problem (for a specified refractive index, type of the particle distribution function, and grid of radius ranges), the spectral AOD values are used to calculate the particle distribution function (dV/dr) or (dS/dr).

2nd step. In the distribution (dV/dr) thus obtained we select its part referring to the fine fraction, and, for it, calculate the total particle volume (V^{f}) through integration.

3rd step. We consider the regression interrelation between particle volumes in the fine fraction (V^{f}) and the τ^{f} values, calculated by empirical method (EM). The interrelation thus

obtained (Fig. 4a) is used to select the parameters of a linear regression equation which makes it possible to calculate the component t^{f} according to the particle volumes $V^{f''}$. (2) "The inverse problem on retrieving the distribution functions (dS/dr) was solved using iteration algorithm of M.A. Sviridenkov [Sviridenkov, 2001], modified from Twitty algorithm [Twitty, 1975]. The particle distribution was assumed to be lognormal, and the refractive index was assumed to have the real part of 1.5 and the imaginary part of 0. In the calculations we used the following radius grid: 0.09-0.13-0.17-0.21-0.25-0.29-0.33-0.37-0.41-0.45-0.49-0.53-0.59-0.65-0.81-0.99-1.21-1.59-1.81-2-2.5-3 µm".

Line 256. Thank you, corrected!

Interannual and seasonal variations in aerosol optical depth of the atmosphere in two regions of Spitsbergen Archipelago (2002-2018)

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Abstract. In this work hourly averaged sun photometer data from the sites Barentsburg and Ny-Ålesund, both located in Spitsbergen in the European Arctic, are compared. Our data set comprises the years 2011 to 2017. We found for more turbid 10 periods (aerosol optical depth $\tau_{0.5} > 0.1$) that typically Barentsburg is more polluted than Ny-Ålesund, especially in the short wave spectrum. However, the diurnal variation of AOD is highly correlated. Next, τ was divided into a fine and coarse mode. It was found that generally the fine mode aerosol optical depth dominates and also shows a larger interannual as inner annual variation. The fine mode optical depth is in fact larger in spring during the Arctic Haze period. Overall the aerosol optical depth seems to decrease, although this is not statistically significant. 15

1 Introduction

The studies of the character and causes of variations in all components of climate system, including the aerosol composition of the atmosphere, became more urgent with regard to climate change [IPCC, 2013]. Atmospheric aerosol plays an important role in the processes of solar radiative transfer and exchange by different substances (and, in particular, pollutants) between

- the continents and ocean [e.g. Kondratyev et al., 2006]. As compared to gases, aerosol is characterized by a complex 20 physicochemical composition and stronger variation of concentration and radiative impact. Of the various aerosol characteristics, the observations of aerosol optical depth (AOD) of the atmosphere are most widespread and carried out at the international and national networks of stations using sun photometers (see, e.g., [WMO, 2005; Holben et al., 1998]). The AOD represents the extinction of radiation, integrated over atmospheric column, and can be considered as
- an optical equivalent of the total aerosol content. 25

One of the main aerosol climatology problems is to determine the specific features of interannual and seasonal variations in different regions. However, under the conditions of changing climate system, a time series 10 years (or even 20 years) long may turn out to be insufficient to identify correctly the tendencies or periodicities in variations of aerosol characteristics. First observations of spectral AOD in the Arctic zone were carried out about 40 years ago [Shaw, 1982; Freund, 1983;

Radionov and Marshunova, 1992]; however, they become regular in character only in the early 2000s, after the development 30



of photometric observations at continental stations. A comprehensive overview of atmospheric AOD in the Polar Regions has been presented by C. Tomasi [Tomasi et al., 2012; 2015].

Studies by various authors showed that the Arctic atmosphere is affected appreciably by outflows of aerosols of different types (smoke, industrial, sulfate, organic) from Eurasia and North America. The most powerful effect is due to smoke from forest

- fires that cover large areas of boreal zone [e.g. Chubarova et al., 2012; Sitnov et al., 2013; Zhuravleva et al., 2017]. Long-range transports of smoke plumes lead to a considerable pollution of the Arctic atmosphere [Stohl et al., 2006; Stone et al., 2008; Eck et al., 2009; Vinogradova et al., 2015]. These episodes are short-term (1-3 days) and rare because they depend on the product of probabilities of two independent events: (a) the fire itself in any area of boreal zone, and (b) the fact that the trajectory of air transport from a fire center arrived exactly at a given region of the Arctic.
- 40 In addition to smokes, anthropogenic and other types of fine aerosol also outflow from midlatitudes. In contrast to forest fires, the sources of these aerosols operate almost all the time and are distributed over the entire territory of human life activity. A somewhat larger concentration of anthropogenic aerosol in densely populated regions of Europe was observed in the past century; however, recently the industrial emissions have been stabilized or reduced in this area [Tørseth et al., 2012; Li et al., 2014; Zhdanova et al., 2019].
- 45 An AOD increase may also be associated with volcanic activity. In the period of time considered here, there were no large eruptions (like Pinatubo volcano in 1991), having a global effect. The effects of less powerful volcanic eruptions on the Arctic atmosphere are short-term to mid-term (some weeks in duration) and comparable to those due to smokes from forest fires. For instance, AOD increase on Spitsbergen was observed after the eruptions of volcanoes Kasatochi (August 2008) and Sarychev (July 2009) [Hoffmann et al., 2010; Toledano et al., 2012].
- 50 The effects of pollutant outflows on the Arctic atmosphere intensify in late winter early spring. The temperature inversions in this season lead to the formation and accumulation of aerosol in separate layers of the troposphere (the Arctic Haze Phenomenon) [e.g. Shaw, 1995; Quinn et al., 2007; Tomasi et al., 2015]. In the recent decade increasingly more work was published analyzing the multiyear AOD variations in different regions, either

based on spectral [Zhdanova et al., 2019; Chubarova et al., 2016; Putaud et al., 2014; Li et al., 2014; Xia, 2011; Michalsky et

- al., 2010; Sakerin et al., 2008a; Weller et al., 1998] or integrated (actinometric) [Gorbarenko and Rublev, 2016; Plakhina et al., 2009; Ohvril et al., 2009] measurements of solar radiation. In most regions of Eurasia the AOD shows a decreasing tendency following 1995. The negative AOD trend is associated with decreasing anthropogenic load and the absence of powerful volcanic eruptions analogous to Pinatubo (July 1991) and El Chichón (March April 1982). Of particular interest is the dynamics of aerosol constituent of the atmosphere in the Arctic zone where there are the largest climate changes [IPCC,
- 60 2013]: increased temperature and prolonged warm period, reduction of sea-ice area, changes in the circulations, etc. The archipelago of Spitsbergen in the European Arctic is a special region because it is strongly influenced by the warm West Spitsbergen Current. Hence, for the given geographical latitude the West Coast of Spitsbergen faces warm and moist conditions. (Walczowski and Piechura, 2017). Moreover, Spitsbergen currently faces a pronounced winter warming of about 2 degrees per decade, which can partly be explained by more efficient advection from the Atlantic Ocean (Isaksen et al., 2015;

65 Dahlke and Maturilli, 2017). For this reason aerosol properties over Spitsbergen may not be directly comparable to aerosol observations at other Polar sites. On the other hand, we may see aerosol properties at Spitsbergen now which are more typical for the Arctic in the future in warmer, more marine conditions.

For Spitsbergen an analysis of aerosol properties for separate periods was already performed before [Herber et al., 2002; Glantz et al., 2014; Chen et al., 2016; Pakszys and Zielinski, 2017]. In this work, we discuss the AOD measurements in 2002-2018 in

70 two regions of Spitsbergen: Ny-Ålesund and Barentsburg. Based on the multiyear observation time series, we considered the following issues: (a) differences in AOD between neighboring regions; (b) choice of the method for extracting the contributions of two (fine and coarse) AOD components; (c) seasonally average AOD variations during the polar day; and (d) the specific features of the interannual AOD variations during two periods of measurements (2002-2010 and 2011-2018).

2 Instruments, methods, and observational data

75 2.1 Characterization of instruments and regions of measurements

Historically, observations of the spectral AOD $\tau^{a}(\lambda)$ on Spitsbergen Archipelago are carried out at three closely lying scientific stations (in the order of decreasing latitude): Ny-Ålesund (78°54'N, 11°53'E), Barentsburg (78°04'N, 14°13'E), and Hornsund (77°00'N, 15°34'E). The distances from Barentsburg to Ny-Ålesund and Hornsund are 110 and 120 km respectively.

Measurements of atmospheric AOD in the scientific settlement Ny-Ålesund (population of about 100 residents during summer)
have been performed since 1991. At the first stage (1991-1999) the observations were performed in separate periods of the year (polar day, polar night) using photometers of different types (Sun, Moon, Star). The results of those studies were considered in [Herber et al., 2002]. Here, we analyze the AOD variations in a later period when measurements became regular and homogeneous in character. The main characteristics of sun photometers (SP1A and SP2H), used in measurements, are

85 The sun photometer in Ny-Ålesund is located just south of the settlement in about 10m asl on the BSRN radiation field. The temporal resolution of the data is 1 minute. The instruments are regularly calibrated in Izaña /Tenerife. The air masses for aerosol and for ozone were considered according to the formulas by Kasten and Young (1989) and the WMO No 183 report (2008), respectively. A cloud screening similar to Alexandrov et al. (2004) has been performed.

90

presented in Table 1.

Table 1. Characteristics of Sun-photometers and data volume in Ny-Ålesund and Barentsburg

Scientific stations	Sun-	Angle of view,	Range of spectrum // number of	Number of hours (days) of
	photometers	degrees	spectral channels	measurements
Ny-Ålesund (AWI,	SP1A,	1	0.37-1.06 μm // 13	7520 (1130)
Germany)	SP2H		0.37-1.05 μm // 12	
Barentsburg (AARI and	SPM,	< 2,5	0.34-2.14 μm // 11	1732 (350)
IOA SB RAS, Russia)	SP-9	< 2	0.34-2.14 μm // 13	

In 2011 the monitoring of aerosol characteristics (including AOD [Sakein et al., 2018a]) was organized in the Russian Scientific Center, located in the southwestern part of Barentsburg settlement on the coast of Grønfjorden Gulf. Products of coal-mining industry and thermal power plant, located at distance of about 1 km, may influence the aerosol characteristics in Barentsburg (population of about 500 residents).

- 100 The atmospheric AOD was initially measured using SPM portable sun photometer [Sakein et al., 2013] at an altitude of 65 m above sea level. In 2015 it was changed to SP-9 photometer with automatic Sun tracking system (the instrument was installed at the altitude of 74 m). The base set of the wavelength channels comprises the interference filters with the passband maxima at the wavelengths: 0.34, 0.37, 0.41, 0.50, 0.55, 0.67, 0.77, 0.87, 1.04, 1.25, 1.55, and 2.14 μm. Still another wavelength channel (0.94 μm) is used to determine the total water vapor content of the atmosphere.
- 105 The method for calculating the spectral AOD [Kabanov and Sakerin, 1997; Kabanov et al., 2009] includes accounting for the spectral transmission functions of light filters, and Rayleigh scattering and absorption by atmospheric gases: H₂O, O₃, CO₂ and others. The absorption is calculated using the databases of spectral line parameters HITRAN-2000 (http://www.hitran.com), models of continual absorption MT_CKD (http://rtweb.aer.com/continuum_code.html) and vertical gas distribution AFGL [Anderson et al., 1986]. Water vapor absorption is accounted for using real water vapor contents,
- 110 measured in the wavelength channel of 0.94 μ m.

Total amount of data (hours and days of measurements), which were used in the AOD analysis in two regions, is presented in Table 1. Seasonal and interannual AOD variations were analyzed for the period of polar day (March-September). First the hourly average AOD values were used to calculate the averages for each day of measurements, then the monthly averages were calculated, based on which the averages for each year (or, more specifically, from March to September) were determined.

115 For brevity, they will be called Daily, Monthly, and Annual AOD. The interannual variations in AOD were estimated in two variants: (a) according to the average values in the measurement period (Annual AOD); and (b) according to the averages in periods of spring maximum and autumn minimum of AOD.

2.2 Data comparison and preparation of observation time series

A comparison of observations using two photometers may be of interest for intercalibration of the instruments, i.e., estimating instrumental-methodic AOD determination errors (for the Polar Regions e.g. Mazzola et al. 2011). If the measurements are separated in space (as in the given case), the difference in the data makes it possible to estimate the local AOD inhomogeneities

- 125 (Sakerin et al. 2010), caused by anthropogenic or natural factors: local weather conditions, type and state of the underlying surface, orography, and the effect of industrial or other sources of aerosol. It should be noted that we already compared earlier the AOD measurements at the neighbouring stations on Spitsbergen Archipelago, i.e., Hornsund and Ny-Ålesund [Toledano et al., 2012; Pakszys and Zielinski, 2017]. Comparison of time independent measurements showed that the average difference in the annual and seasonal AOD values at the wavelength of
- 130 0.55 µm does not exceed 0.01-0.02. Large differences in AOD between these stations occur in periods of Arctic haze and outflows of smokes from forest fires, which are differently manifested in these two regions. However, a comparison of monthly averages may introduce a bias due to different data availability at the involved sites. In contrast to the above-mentioned works, we compared quasi-synchronous (nearly time coincident) AOD measurements in
- the neighbouring regions in 2011-2017 (see [Kabanov et al., 2018] for more details). The data of the SP1A (Ny-Ålesund) and
 SPM (Barentsburg) photometer observations were used to calculate the hourly average AOD values. Then, the datasets from
 the two regions were "juxtaposed" provided that the times of the AOD measurements differed by no more than one hour.
 Comparison of measurements with the two photometers showed a large dispersion of the data under the conditions of strong
 atmospheric turbidities, namely, during outflow of smoke plumes from forest fires and in the Arctic Haze situations. Due to
- large spatial inhomogeneity of these structures, AOD, measured in two regions, may strongly differ, making the comparison incorrect. Therefore, further analysis was performed for usual situations, when τ^a (0.5 µm) < 0.2. Figure 1 illustrates the regression relation between τ^a (0.5 µm) measurements in the neighbouring regions of Spitsbergen.

A comparison of the statistical characteristics showed that the average AOD values are a little larger in Barentsburg than in Ny-Ålesund [Kabanov et al., 2018]. The maximum difference in AOD is observed in the shortwave part of the spectrum (0.38 μ m), $\Delta = \tau^a$ (SPM) – τ^a (SP1A) = 0.024; while in IR range (0.87 μ m) the difference decreases to $\Delta = 0.005$. This feature is

145 real despite the decreasing AOD at longer wavelengths and indicates that fine aerosol is more abundant in the atmosphere of Barentsburg. At the same time, we note that the AOD differences are minor (comparable with uncertainty of determining AOD – about 0.01-0.02 [Kabanov et al. 2009; Sakerin et al., 2013]), and the interdiurnal AOD variations in the two regions are coordinated in character (correlation coefficients are 0.83-0.89). Comparison of quasi-synchronous AOD measurements in Barentsburg and Hornsund gave close results [Kabanov et al., 2018]: $\Delta = 0.004 - 0.024$, the correlation coefficients are 0.71150 0.81. Hence, observations in the neighbouring regions on Spitsbergen are quite compatible and identically reflect the specific features of the AOD variations.

The joint use of results from AOD monitoring in the neighbouring regions makes it possible to control the reliability of information, as well as to identify the specific features of AOD variations not only for a specific site, but for the region as a whole. The results of the observations in each of the regions have their own advantages. The advantage of the data from

- 155 Barentsburg (SPM / SP-9 photometers) is a wider range (0.34-2.14 µm) of the spectral measurements and the possibility to separate the contributions from two AOD components, using an empirical method (see subsection 2.3). The valuable feature of the data from Ny-Ålesund is a longer AOD observation time series. However, different errors have been accumulated in these data for the long period of measurements. A simple exclusion of all suspect AOD measurements was undesirable because for analysis of multiyear variations it was necessary to keep the observation time series as long as
- 160 possible. Taking this circumstance into account, the multiyear observation time series was prepared for sorting out or correction of suspect AOD values [Kabanov et al., 2019a]. The initial dataset was processed to remove the data in which short-term bursts or rapid AOD variations were observed, as well as the distortions to smoothness of the wavelength dependences $\tau^a(\lambda)$. Owing to a certain redundancy of the number of spectral channels, we could identify false measurements and select most reliable data.

2.3 Fine and coarse AOD components

165 The attenuation of radiation by atmospheric aerosol varies as a function of wavelength, depending on sizes and refractive index of aerosol particles. To characterize the AOD, measured at different wavelengths, the Ångström formula is widely used:

$$\tau^{a}(\lambda) = \beta \cdot \lambda^{-a}, \qquad (1)$$

where β and α are the approximation parameters of the spectral dependence of AOD; β is the turbidity coefficient, which is close in value to AOD at the wavelength of 1 μ m; and α is the selectivity exponent (power-law decay).

- 170 Numerous studies in different regions and atmospheric conditions showed that the formula (1) does describe well the wavelength dependence $\tau^a(\lambda)$ in the main range (0.34 1 µm) of AOD measurements. [Angstrom, 1964; Shifrin, 1995; Eck et al., 1999; Cachorro et al., 2000; Schuster et al., 2006]. At the same time, the use of this formula has limitations and disadvantages, requiring an explanation.
- First, the Ångström formula becomes unsuitable for describing the wavelength behavior of AOD in the atmospheric "transparency windows" in the wavelength range of $1 - 4 \mu m$. This is because the power-law dependence (1) stems from the combined action of fine and coarse aerosol fractions, which have different spectral properties. Extinction of radiation by small particles ($2\pi r/\lambda < 1$) is dominant in the visible region of spectrum; however, it rapidly decays with the growing wavelength and becomes insignificant in the region beyond of 1 μm . Extinction of radiation by coarse aerosol barely changes with the wavelength and becomes predominant in the IR range. Mie calculations and experimental data [Sakerin and Kabanov, 2007a;
- 180 Sakerin et al., 2008b] confirm that the power-law AOD decay gradually goes over into almost neutral dependence. Therefore, $\tau^a(\lambda)$ in a wider wavelength range is better to represent by a sum of two components:

$$\tau^{a}(\lambda) = \tau^{c} + \tau^{f}(\lambda) = \tau^{c} + m \cdot \lambda^{-n}, \qquad (2)$$

where τ^c is the constant (wavelength independent) coarse AOD component; $\tau^f(\lambda)$ is the selective fine component; m and n are the approximation parameters of the spectral dependence of $\tau^f(\lambda)$ (they are similar to the parameters β and α of the Ångström

185 formula)

Second, the Ångström parameters do not allow one to interpret the causes for AOD variations unambiguously. An increase/decrease in the exponent α is sometimes unjustifiably attributed only to changes in fine aerosol. In fact, the exponent α conceals the actions of a few factors. The wavelength dependence of AOD is indeed determined by the optical properties of fine component $\tau^{f}(\lambda)$. Both the sizes and refractive index of small particles influence the degree of wavelength decay of AOD

(and values of the parameters n and α). But, precisely what caused changes in the selectivity of AOD is almost impossible to determine without the use of additional information like e.g. aerosol in-situ measurements.
 The next uncertainty factor is that the exponent α depends on the *relationship* of the optical depths of fine and coarse aerosol (τ^f_λ/τ^c). That is, α may increase both due to growing content of fine aerosol, and to decreasing content of coarse aerosol.

The presence of the interrelation between α and $(\tau_{\lambda}^{f}/\tau^{c})$ is illustrated in Fig. 2. The correlation coefficient between α and ln

195 $[(\tau_{\lambda}^{f} / \tau^{c}) + 1]$ is statistically significant and equal to 0.68 (confidence level P < 0.0001).

We also note that the component τ^c , which influences the exponent α , is tightly related and has close values to the second Ångström parameter [Sakerin and Kabanov, 2007a, b]: $\tau^c \approx \beta$. A consequence of this is that the parameters α and β are themselves correlated.

Thus, the use of the Ångström parameters in the analysis of AOD variations is ambiguous and may lead to erroneous

- 200 conclusions. It is more preferable to consider the specific features of variations in two AOD components: $\tau^{f}(\lambda)$ and τ^{c} . In addition to different sizes and spectral properties, fine and coarse aerosol fractions differ in the origins of particles and their transformation in the atmosphere. Fine (sulfate, organic, etc.) aerosol is formed in the atmosphere as a result of various photochemical and microphysical processes [Kondratyev et al., 2006]. The lifetime of fine aerosol in the troposphere is a few days; therefore, it can be transported long distances (hundreds and thousands of kilometers) away. The main source of coarse
- 205 (marine and dust) aerosol is the underlying surface. Because of its short lifetime and small transport distance, coarse aerosol is more local in character and pertains to a specific terrain. The only exceptions are powerful dust outflows in tropical latitudes.

2.4 Methods for determining fine and coarse AOD components

As was already indicated above, in the IR range, the effect of fine aerosol becomes insignificant, and AOD is determined only 210 by the coarse component. Therefore, τ^c can be determined by an *empirical* method (EM), i.e., from minimal or average AOD values, measured in the range of 1.24-2.14 μ m [Sakerin and Kabanov, 2007b; Sakerin et al., 2008b]. Then, the second (fine) component is found as a residual of the total AOD. Usually, it is estimated for the wavelength of 0.5 μ m: $\tau_{0.5}^f = \tau_{0.5}^a - \tau^c$. However, most sun photometers (and in particular the SP1A in Ny-Ålesund) operate in the wavelength range up to 1.05 μ m, making *empirical* method inapplicable. In this case, τ^c and $\tau_{0.5}^f$ can be estimated using calculation methods. For instance, in

- 215 the AERONET system (http://aeronet.gsfc.nasa.gov), $\tau_{0.5}^{f}$ is calculated using the spectral deconvolution algorithm [O'Neill et al., 2003], based on the relationship of spectral AOD, measured in the shortwave part of the spectrum 0.38-1.02 µm. In the work [Kabanov and Sakerin, 2016] we suggested simpler methods for separating the contributions from $\tau_{0.5}^{f}$ and τ^{c} , based on the regression interrelations with the parameters of Ångström formula. In the first regression method (RM1), τ^{c} is estimated using its interrelation with the parameter β (see formula (3) below). In the second method (RM2), the regression
- 220 dependence of $\tau_{0.5}^{f}$ on the parameters α and β (see formula (4)) is used. Comparison of different methods of $\tau_{0.5}^{f}$ (or τ^{c}) estimation for the conditions of the marine and continental (Tomsk) atmosphere showed close results: the average difference of τ^{c} from data of base *empirical* method (EM) does not exceed 0.007 for the standard deviation from 0.006 to 0.024. For the conditions of Arctic region (Spitsbergen), we performed an additional study [Kabanov et al., 2019a], concerning the selection of an optimal method of $\tau_{0.5}^{f}$ (or τ^{c}) estimation. Different methods were tested using SPM photometer measurements

of AOD in Barentsburg. The error of the methods was estimated by comparing the calculated values of $\tau_{0.5}^{f}$ or τ^{c} with the data from base *empirical* method.

Figure 3 illustrates the results of testing two regression methods (RM1 and RM2), based on the interrelations (a) between τ^c and the parameter β , and (b) between $\tau_{0.5}^f$ and the parameters α , β . For the conditions of Spitsbergen, we obtained the following regression equations:

230 RM1: $\tau^c = 0.665 \cdot \beta - 0.0005$ (3) RM2: $\tau_{0.5}^f = (-0.829 + 1.066 \cdot 0.5^{-\alpha}) \cdot \beta$ (4)

Table 2 presents the standard deviations σ and the correlation coefficients R between the calculated (RM1, RM2) and empirical (EM) τ^c values. These results suggest the regression methods make it possible to estimate τ^c with an identical error of 0.007.

235 The disadvantage of the regression methods is that they require a preliminary data accumulation under the conditions of a specific region for determining optimal regression coefficients in equations (3) and (4) [Kabanov and Sakerin, 2016]. Of course, the error of the regression methods may increase if aerosol characteristics strongly differ from those typical for the region and do not correspond to the selected regression coefficients.

Therefore, in addition to the regression methods, we considered the applicability of another two methods of $\tau_{0.5}^{f}$ estimation, 240 based on the results of solving the inverse problem, namely: retrieval of particle sizes from measurements of spectral AOD. The inversion method 1 (IM1) is based on the interrelation between $\tau_{0.5}^{f}$ and volume or cross section of particles of fine aerosol.

This method is implemented in the following steps:

1st step: Based on any known method of solving the inverse problem (for a specified refractive index, type of the particle distribution function, and grid of radius ranges), the spectral AOD values are used to calculate the particle distribution function

 $245 \quad (dV/dr) \text{ or } (dS/dr).$

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2nd step: In the distribution (dV/dr) thus obtained we select its part referring to the fine fraction, and, for it, calculate the total particle volume (V^{f}) through integration.

3rd step: We consider the regression interrelation between particle volumes in the fine fraction (V^f) and the t^f values, calculated by empirical method (EM). The interrelation thus obtained (Fig. 4a) is used to select the parameters of a linear regression equation which makes it possible to calculate the component t^f according to the particle volumes V^f.

The inversion method 2 (IM2) is implemented by solving first the inverse problem, and then the direct problem of the aerosol optics: (a) as in IM1, the spectral AOD values are used to retrieve the distribution functions (dS/dr); and (b) based on the (dS/dr) values, $\tau_{0.5}^{f}$ is calculated for the size range of fine aerosol.

The inverse problem on retrieving the distribution functions (dS/dr) was solved using iteration algorithm of M.A. Sviridenkov

- 255 [Sviridenkov, 2001], modified from Twitty algorithm [Twitty, 1975]. The particle distribution was assumed to be lognormal, and the refractive index was assumed to have the real part of 1.5 and the imaginary part of 0. In the calculations we used the following radius grid: 0.09-0.13-0.17-0.21-0.25-0.29-0.33-0.37-0.41-0.45-0.49-0.53-0.59-0.65-0.81-0.99-1.21-1.59-1.81-2-2.5-3 μm.
- Applicability of inversion methods was estimated for a few variants: (a) for different wavelength intervals of AOD (0.34-2.14 µm; 0.38-0.87 µm; 0.38-1.02 µm; and 0.34-1.02 µm); (b) for the distribution functions (dS/dr) and (dV/dr); and (c) for different radius boundaries of particles of fine fraction (0.1-0.5 µm and 0.1-0.45 µm). Figure 4 presents examples of interrelations: (a) between $\tau_{0.5}^{f}$ (EM) and calculated values of particle volume V^{f} ; and (b) between $\tau_{0.5}^{f}$ values, determined using base (EM) and inversion (IM1) methods. The calculations in this case were performed for the wavelength range of AOD 0.38-1.02 µm and particle radius range of 0.1-0.5 µm.
- Analysis of application of IM1 and IM2 [Kabanov et al., 2019a] showed that the $\tau_{0.5}^{f}$ determination error decreases by about a factor of 1.5 when the AOD is used in a wide (0.34-2.14 µm) wavelength range. However, for the narrower wavelength range of the SP1A photometer (0.38-1.02 µm) the $\tau_{0.5}^{f}$ calculation error is comparable with results from regression methods (see Table 2). That is, the relative errors of the τ^{c} and $\tau_{0.5}^{f}$ calculations for the mean conditions of Barentsburg ($\tau_{0.5}^{a} = 0.086$ [Sakerin et al., 2018a]) are 30% and 11% respectively.

270 The IM1 method was chosen for a subsequent use. Despite a more complicated procedure of its calculations, IM1 is more sensitive to aerosol variations, which is indicated by the highest correlation coefficient between $\tau_{0.5}^{f}$ (IM1) and data from base

(EM).

There may be a question as to why after retrieval of aerosol size distribution we nonetheless consider the seasonal and interannual variations in optical characteristics: τ^c and $\tau_{0.5}^f$? Analysis of disperse composition of aerosol is a more complex

- and non-unique problem because it is necessary to consider the transformation of two aerosol fractions, which are described by a few parameters: shapes and widths of distributions for each fraction, separation boundary, and effective particle radii. Moreover, an uncertainty remains about the values of these parameters because of the priori specified aerosol refractive index. In this work, we pursued a simpler task: to determine the character and magnitude of variations in aerosol optical characteristics. In this case, instead of many microstructure parameters, it is sufficient to analyze their more compact optical images in the form of two components, of and of
- 280 image in the form of two components, τ^f and τ^c .

Parameter	RM1	RM2	IM1	IM2
σ	0.007	0.007	0.007	0.008
R	0.819	0.963	0.967	0.953

Table 2. Estimates of applicability of different methods (RM1, RM2, IM1, IM2) of the τ^c (or $\tau^{f}_{0.5}$) calculation

285 **3. Discussion of the results**

Current climate change and environmental transformation influence the regularities of variations in aerosol characteristics to some degree. Because of the deficit of its own aerosol sources in the Arctic zone, an important role in AOD variations is played by outflows of smoke, anthropogenic and volcanic aerosol from midlatitudes. The frequency of these outflows in particular months and years determines the specific features of seasonal dynamics of AOD in Arctic regions and magnitude of interannual oscillations.

3.1 Interannual variations

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The highest atmospheric turbidities in the region of Spitsbergen were observed on July 10, 2015 and on May 2-3, 2006 (Fig. 5). Daily AOD ($0.5 \mu m$) in these cases reached 0.82 and 0.6, about an order of magnitude lager than multiyear averages. After passing to monthly AOD values, the effect of these short-term turbidities decreased to 0.192 in May 2006 and 0.152 in July 2015. Trajectory analyses of air mass motion showed that the extreme AOD values in July 2015 were due to long-range transport of smokes from forest fires in Alaska [Sakerin et al., 2018a, Pakszys and Zielinski, 2017; Markowicz et al., 2016]. We also considered in detail the second anomalous situation (in May 2006) [e.g. Myhre et al., 2007; Stohl et al., 2007], associated with the outflow of smoke aerosol from agricultural fires in the Eastern Europe.

Episodes with high atmospheric turbidities were also observed in June 2003, March and August 2008, in April and August

300 2009, and in April 2011. The AOD values in these periods of time had already been analyzed by many authors [Toledano et al., 2012; Glantz et al., 2014; Tomasi et al., 2015; Chen et al., 2016; Pakszys and Zielinski, 2017]. Independent of the causes for these short-term turbidities (Arctic haze, outflows of smoke or volcanic aerosol), they enhance not only the monthly, but also annual AOD values.

The above-mentioned high-turbidity episodes (2006, 2008, 2009, 2015) were reflected partly in annual AOD oscillations 305 (Fig. 6). Moreover, a maximum appeared in the interannual variations in 2011-2012. This maximum was due not to extreme 1-3-day AOD bursts, but to stronger turbidities as compared to the neighbouring years.

The annual AOD maxima occur with the average periodicity of about three years. When high-turbidity episodes are eliminated (see dashed line in Fig. 6), certain maxima disappear; however, the general character of the AOD oscillations remains unchanged. Among these maxima, the highest AOD value in 2003 seems suspect. This annual AOD value cannot be considered as representative because of short period of observations (4 days in March and 5 days in May-June) in this year.

In addition to oscillations, a tendency for a minor AOD decrease can be discerned in the multiyear variations. This is also indicated by a comparison of AOD characteristics in two periods of observations: the average AOD (0.5 μ m) in 2011-2018 decreased by 0.013 relative to 2002-2010 (Fig. 5). However, this decreasing AOD tendency is not statistically significant. The statistical estimates of $\tau_{0.5}^{f}$ and τ^{c} variations in spring and fall periods in these two regions (Fig. 7) also revealed no trend

315 component.

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From the statistical characteristics (Table 3) and Fig. 7 it follows that AOD and its interannual variations are determined mainly by fine aerosol: the SD values and the range <Min – Max> are a factor of 2-3 larger for $\tau_{0.5}^{f}$ than τ^{c} . The relative variations $\tau_{0.5}^{f}$ and τ^{c} are about the same: their variation coefficients V are 14-29%. Neither AOD component shows a clear predominance of variation coefficients. For instance, in the period of 2011-2018, the variation coefficients were larger for $\tau_{0.5}^{f}$ than τ^{c} (25 and 20%) in Ny-Ålesund and with a reverse relationship (14 and 23%) found in Barentsburg.

The interannual oscillations in the Ångström exponent can be considered as minor: the variation coefficients for α are 13-15%. The average α and the total variability range (from 1 to 1.7) are in the regions of values characteristic for the continental midlatitude atmosphere and are larger than in the marine atmosphere [Sakerin et al., 2008b; 2018b]. These values of the Ångström exponent are because the ratio ($\tau_{0.5}^{f}/\tau^{c} = 2.6-2.9$) and the relative contribution of fine component ($\tau_{0.5}^{f}/\tau_{0.5}^{a} = 0.73$ -

325 0.75) are close to continental values. As an example, we present multiyear data in boreal zone of Siberia in spring (smokefree) period [Kabanov et al., 2019b]. The average AOD values in Siberia are about two times larger ($\tau_{0.5}^{f} = 0.105$, $\tau^{c} = 0.036$) than in Ny-Ålesund, and the ratio ($\tau_{0.5}^{f}/\tau^{c}$) and the exponent α are almost the same: ($\tau_{0.5}^{f}/\tau^{c}$) = 2.92 and α = 1.43. Table 3. Statistical characteristics of Annual AOD: average, minimal (Min), maximal (Max) values, standard deviations (SD),
and variation coefficients (V); values for Ny-Ålesund (2002-2018) in the first row, for Ny-Ålesund (2011-2018) in the second row; and for Barentsburg (2011-2018) in the third row

Mean	SD	Min	Max	V, %
0.067	0.017	0.04	0.10	25
0.059	0.012	0.04	0.08	20
0.080	0.007	0.07	0.09	10
0.050	0.013	0.02	0.07	26
0.044	0.011	0.02	0.06	25
0.058	0.008	0.04	0.07	14
0.017	0.005	0.01	0.03	29
0.015	0.003	0.01	0.02	20
0.022	0.005	0.01	0.03	23
1.41	0.19	1.03	1.71	13
1.44	0.21	1.11	1.71	15
1.24	0.19	1.01	1.60	15
0.026	0.008	0.016	0.040	31
0.022	0.006	0.016	0.034	27
0.035	0.005	0.025	0.042	14
	Mean 0.067 0.059 0.080 0.050 0.044 0.058 0.017 0.015 0.022 1.41 1.44 1.24 0.026 0.022 0.035	Mean SD 0.067 0.017 0.059 0.012 0.080 0.007 0.050 0.013 0.044 0.011 0.058 0.008 0.017 0.005 0.015 0.003 0.022 0.005 1.41 0.19 1.44 0.21 1.24 0.19 0.026 0.008 0.022 0.006 0.035 0.005	Mean SD Min 0.067 0.017 0.04 0.059 0.012 0.04 0.080 0.007 0.07 0.050 0.013 0.02 0.044 0.011 0.02 0.0458 0.008 0.04 0.017 0.005 0.01 0.015 0.003 0.01 0.015 0.003 0.01 0.022 0.005 0.01 1.41 0.19 1.03 1.44 0.21 1.11 1.24 0.19 1.01 0.026 0.008 0.016 0.022 0.006 0.016	MeanSDMinMax0.0670.0170.040.100.0590.0120.040.080.0800.0070.070.090.0500.0130.020.070.0440.0110.020.060.0580.0080.040.070.0170.0050.010.030.0150.0030.010.020.0220.0050.010.031.410.191.031.711.240.191.011.600.0260.0080.0160.0400.0350.0050.0250.042

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From Figs. 6 and 7 it is clearly seen that AOD in Barentsburg is almost always higher than in Ny-Ålesund. The average excess of annual AOD is (see rows 2 and 3 in Table 3): $\Delta \tau_{0.5}^a = 0.02$, $\Delta \tau_{0.5}^f = 0.012$, $\Delta \tau^c = 0.008$. This result indicates that a little more anthropogenic (fine) and coarse aerosols are contained in the atmosphere of Barentsburg (see also Figs. 1 and 5).

Variations in annual AOD in Ny-Ålesund and Barentsburg are coordinated in character. Oscillations in fine component 340 sometimes show opposite changes in the two regions, such as in spring 2013 (see Fig. 7). Different behaviors of $\tau_{0.5}^{f}$ may be because observation time series are inhomogeneous in each region due to clouds or because AOD are measured at different times.

3.2 Specific features of seasonal variations

The most common regularity of the seasonal AOD behavior at midlatitudes is the spring (and sometimes also summer)

345 maximum and fall minimum [e.g., Sakerin et al., 2015; Chubarova et al., 2014; Holben et al., 2001]. The primary causes for this AOD behavior are the annual cycle in the Sun's declination meaning a return of sunlight and possibly a longer aerosol life-time over the frozen ocean. The springtime increases in insolation and temperature trigger a few processes: (a) snow cover evaporates and melts; (b) the atmosphere is enriched by different deposition products, accumulated over the winter, (c) primary (marine, soil) aerosol starts to come from the underlying surface; and (d) photochemical processes of production of in situ 350 aerosol in the atmosphere and emission of organic aerosol are activated [e.g., Kondratyev et al., 2006].

The seasonal AOD dynamics in the Arctic zone is analogous to midlatitudes: springtime maximum and a decay toward fall [e.g., Toledano et al., 2012; Tomasi et al., 2015; Sakerin et al., 2018]. This AOD behavior is because of similar annual rhythms of both own aerosol sources in the Arctic and long-range aerosol transports from midlatitudes.

Figure 8 shows the seasonal variations in monthly AOD for two regions in Spitsbergen. The seasonal AOD behaviours in Ny-Ålesund were similar in character in 2002-2010 and 2011-2018. The difference is that AOD values in March-June have decreased by ~0.02 in recent 8 years. At the same time, monthly AOD in the second half of polar day (July-September) remained at the same level of low values 0.05-0.06. As a result, the seasonal AOD decrease in 2011-2018 became less pronounced: the relative amplitude had been 30% (versus 55% in the period of 2002-2010).

Seasonal AOD variations in Barentsburg are characterized by an additional summer maximum in July-August. Despite this
difference, common factors in AOD variations in the two regions are nonetheless predominant. Analysis of interrelation
between AOD values, measured in Ny-Ålesund and Barentsburg (Fig. 9), showed quite a high (0.90) correlation coefficient.
Hence, the synoptic, seasonal, and interannual AOD oscillations are largely coordinated in character.

Observation time series in the two regions were compared to clarify the causes for the summertime AOD maximum in Barentsburg. The increased AOD values in July and August were found to be due to the situations with smoke outflows (and,

- 365 in particular, on July 10, 2015 [Sakerin et al., 2018a]). Of the total number of measurements, percentage of smokecontaminated measurements turned out to be larger in Barentsburg than in Ny-Ålesund. A few rare, but powerful outflows of smoke aerosol have led to an increase in the Monthly $\tau_{0.5}^{f}$ (Fig. 10) and $\tau_{0.5}^{a}$ (Fig. 8) values and distorted the natural seasonal variations. After the events of smoke outflows are eliminated (see dashed lines in Figs. 8 and 10), the seasonal AOD behavior in Barentsburg becomes similar to that in Ny-Ålesund, but with larger (by 0.017, on the average) AOD values. The average
- 370 AOD characteristics for the periods of spring maximum and fall minimum in the two regions are presented in Table 4 and in Fig. 11.

Table 4. Average AOD characteristics (± SD) in Ny-Ålesund and Barentsburg during spring (April-May) maximum (first row) and fall (August-September) minimum (second row)

Characteristics	Ny-Ålesund,	Ny-Ålesund,	Barentsburg, 2011-	
	2002-2018	2011-2018	2018 (No smoke)	
$ au_{0.5}^{a}$	0.081±0.03	0.070±0.015	0.086±0.012	
	0.052±0.023	0.052±0.015	0.070 ± 0.022	
$ au_{0.5}^{f}$	0.062±0.024	0.054±0.012	0.063±0.012	
	0.038±0.019	0.038±0.013	0.048 ± 0.02	
τ^{c}	0.019±0.007	0.016±0.005	0.022±0.009	
	0.014±0.004	0.014±0.003	0.022±0.011	
α	1.43±0.23	1.47±0.22	1.33±0.28	
	1.38±0.26	1.35±0.30	1.17±0.36	
β	0.031±0.01	0.026 ± 0.007	0.035±0.008	
	0.02 ± 0.008	0.02 ± 0.005	0.032±0.012	

From Fig. 10 and Table 4 it can be seen that the fine component makes the major contribution to the formation of the seasonal AOD behavior: the average $\tau_{0.5}^{f}$ values decrease from spring toward fall in the two regions by almost the same amount of 0.015-0.016. Also, modal (most probable) $\tau_{0.5}^{f}$ values vary in a similar way (Fig. 12a). The $\tau_{0.5}^{f}$ mode from spring toward fall shifts from 0.07 to 0.03 in Barentsburg and from 0.05 to 0.03 in Ny-Ålesund. Average (Fig. 10) and modal (Fig. 12b) values of coarse AOD component remain almost unchanged during the polar day: Monthly τ^{c} values are about 0.015 in Ny-380 Ålesund and 0.022 in Barentsburg.

Seasonal decrease of $\tau_{0.5}^{f}$ from spring toward fall leads to changes in the ratio ($\tau_{0.5}^{f}/\tau^{c}$) and spectral AOD dependence (Fig. 11): the slope of the spectral AOD dependence and the Ångström exponent become a little smaller. The relative contribution of fine aerosol to AOD ($\tau_{0.5}^{f}/\tau_{0.5}^{a}$) in Ny-Ålesund is 0.77 in spring and 0.73 in fall. In Barentsburg this ratio is a little smaller, 0.73 and 0.69 respectively.

385

4 Conclusions

We present brief results of our study.

It is noted that, to identify the specific features of seasonal and multiyear variations in atmospheric AOD, it is important
 to analyze separately fine and coarse AOD components, having different spectral properties, origins, and lifetimes. As applied

to AOD measurements in Ny-Ålesund, we considered a few methods for estimating the contributions of fine and coarse components, and one of the methods (IM1) is selected for a subsequent use. A comparison with data from the base (EM) showed that the standard deviation of the τ^c and $\tau_{0.5}^f$ calculations is 0.007, and the relative errors are 30% and 11% respectively.

- 395 2. Outflows of fine aerosol of different types from the Eurasian and North American midlatitudes affect appreciably the monthly (and even annual) AOD in the Arctic atmosphere. Outflows of smokes from massive forest and agricultural fires have the strongest effect. The frequency of these episodic outflows in specific years, as well as the frequency of situations of Arctic haze, influence the specific features of seasonal variations and determine the amplitude of interannual AOD variations in the Arctic atmosphere.
- 3. The oscillations in annual AOD in Ny-Ålesund and Barentsburg are coordinated in character and determined by fine aerosol (interannual variations in τ^c are a factor of 2-3 smaller). In the multiyear (2002-2018) variations we revealed a tendency of decreasing AOD, but the trend is statistically insignificant. Annual AOD in Barentsburg is, on the average, 0.02 larger than in Ny-Ålesund, indicating larger contents of both fine ($\Delta \tau_{0.5}^f = 0.012$) and coarse ($\Delta \tau^c = 0.008$) aerosol in the bigger settlement.
- 4. The seasonal AOD variations in Ny-Ålesund are characterized by a decrease from spring toward fall. In the last period (2011-2018) the seasonal AOD decrease became less pronounced. Monthly AOD values decreased from 0.07-0.09 by the amount of ~0.02 in March-June, and remained unchanged (0.05-0.06) in the second half of polar day. In the seasonal AOD variations in Barentsburg there had been an additional summer maximum, caused by a relatively larger influence of smoke outflows. After smoke outflow events are eliminated, the seasonal behaviour becomes analogous to that in Ny-Ålesund, but with (0.017) higher AOD values.
- 410 5. Fine component has the main effect on the formation of seasonal behaviour of AOD. Its relative contribution to AOD ($\tau_{0.5}^{j}$

 $/\tau_{0.5}^{a}$) is 0.77 in spring and 0.73 in fall in Ny-Ålesund (0.73 and 0.69 in Barentsburg). Coarse AOD component during the polar day is almost unchanged, being, on the average, 0.015 in Ny-Ålesund and 0.022 in Barentsburg. The average annual and monthly values of the Ångström exponent α (from 1.2 to 1.5) do not differ from those at the midlatitudes of the continental atmosphere. That large exponents α are because the ratios of the two AOD components ($\tau_{0.5}^{f}/\tau^{c}$) differ little between the Arctic

415 and continental atmosphere.

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Fig. 1. Scatter diagram of AOD measurements in two regions using SP1A (Ny-Ålesund) and SPM (Barentsburg) photometers. Solid line: unconstrained linear regression; dotted line: regression through origin (0,0)



Fig. 2. Correlation dependence between the exponent α and the ratio $(\tau_{0.5}^f / \tau^c)$ according to measurements in Barentsburg. Solid line: unconstrained linear regression; dotted line: regression through origin (0,0)



Fig. 3. (a) Interrelation of τ^c with the parameter β and (b) interrelation of $\tau_{0.5}^f$, calculated using the base (EM) and RM2 methods



Fig. 4. (a) Interrelation between $\tau_{0.5}^{f}$ and particle volume V^{f} and (b) interrelation between $\tau_{0.5}^{f}$ values, calculated using inversion (IM1) and empirical (EM) methods



Fig. 5. Spectral dependences of AOD: at the top shows situations with high aerosol turbidities of the atmosphere; and the bottom shows multiyear averages in two regions



640 Fig. 6. Multiyear variations in Annual AOD in Ny-Ålesund and Barentsburg



Fig. 7. Multiyear variations in $\tau_{0.5}^{f}$ and τ^{c} for the periods of spring maximum and fall minimum of AOD in two regions of Spitsbergen



Fig. 8. Seasonally average $\tau^a_{0.5}$ variations in Ny-Ålesund and Barentsburg



Fig. 9. Interrelation between daily AOD (0.5 μm) values, measured in Ny-Ålesund and Barentsburg (2011-2018). Solid line: unconstrained linear regression; dotted line: regression through origin (0,0)



650 Fig. 10. Seasonal variations in fine and coarse AOD components in Ny-Ålesund and Barentsburg



Fig. 11. Average spectral dependences of AOD in two regions of Spitsbergen (2011-2018) during spring (April-May) maximum and fall (August-September) minimum



Fig. 12. Frequency histograms of (a) $\tau_{0.5}^{f}$ and (b) τ^{c} in spring and fall in two regions of Spitsbergen (2011-2018)





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