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Atmospheric Chemistry and Physics Discussions

# *Interactive comment on* "On measurements of aerosol–gas composition of the atmosphere during two expeditions in 2013 along Northern Sea Route" *by* S. M. Sakerin et al.

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Reply to comments of Reviewer # 1

We thank the reviewer for constructive and valuable comments regarding our paper. We tried to implement your comments as full as possible: the required revisions (additions) are made and questions are answered.

1) Introduction. Additional material is introduced in Introduction section (page 16779, at the end of line 3): After the sentence "The long-range meridional transports of aerosol substance occur predominately during winter and in submicron particle size



range" please add: "For this reason, situations with elevated aerosol turbidity of the atmosphere, known since last century as "Arctic haze", are observed in winter-spring period.

Recent studies (Roiger et al., 2015; Law et al., 2014; Stohl et al., 2013 and references therein) showed that a number of other factors, overlooked or underestimated in previous climate modeling, should be taken into consideration. Primarily, there had been an accelerated development of northern territories, associated with extraction and processing of mineral resources, combustion of petroleum gas in the regions of oil and gas production, and intensification of sea and air transportation. Climate changes (growth in temperature and duration of blocking anticyclones) gave rise to more frequent severe forest fires, leading to formation of large masses of smoke aerosol and its transport to Arctic (e.g., Stohl et al., 2006; Stone et al., 2008; Eck et al., 2009). For instance, in boreal zone of Russia, large-scale events of extreme smoke haze were observed in 2002, 2010, and 2012 (e.g., Chubarova et al., 2012; Gorchakov et al., 2014; Kozlov et al., 2014). Moreover, aerosol-gas (including black carbon) emissions from heating systems in cold period had enhanced due to the development of Arctic.

Thus, ever growing emission of natural and anthropogenic aerosol in subarctic regions is added to far-range aerosol transport from Eurasia in modern period. A consequence of all the above is the change in radiative characteristics of the Arctic atmosphere (including clouds); while sedimentation of absorbing aerosol leads to a reduction in snow surface albedo, temperature rising, and ice and snow melt, i.e., just to those processes the role of which is least studied at present (IPCC, 2013)."

2) Section 2, Table 1. According to Reviewer's comment, additional information is introduced in Table 1. Modified Table 1 is presented in the Supplement (pdf-file).

In the last paragraph on page 16783 (lines 23-28) excluded the first two sentences. The paragraph begins with a revamped offer: "Aerosols were accumulated on filters for 8–10 hours using air pumps (see Table 1)". On page 16784, line 15, and on page

16785, line 3, instead of literature citations we added: (see Table 1).

3) Section 3.1, Table 2: The regions of ground-based observations are already mentioned in the text on page 16785, line 23-26. In accordance with Reviewer's comments, the words "Coastal station" are added in Table 2; they indicate three regions where ground-based observations were performed. Modified Table 2 is presented in the Supplement (pdf-file).

4) Section 4.1:

(a) The regions for calculating the statistics of AOD and microphysical parameters (Ma, Na, Mbc) on the route of RV Akademik Fedorov were identified individually, depending of what data were available in the study regions. In particular, AOD was measured only at the points 2 and 4 of the route, contained in the region A (see Fig. 1). No AOD measurements were performed in the regions K and B due to the presence of clouds. Instead, observations from coastal Arctic stations Barentsburg (region 1) and Polyarka (region 3) were employed for comparison purposes. A sufficient amount of data along the route of RV Professor Khljustin made it possible to perform uniform division of different characteristics with respect to the regions (seas).

In a preliminary analysis of aerosol chemical composition, region A was divided into 3 sub-regions (A1, A2, A3). However, the compositions of separate samples strongly varied, so that no significant differences between sub-regions were found. Large amount of data on the route of RV Professor Khljustin had permitted a uniform division of different characteristics with respect to regions (seas). Because of the association of Sections 5 and 6, we have made changes in Fig. 1: excluded sub-regions A1, A2, A3. (The updated Fig. 1 are presented )

(b) We thank the Reviewer for providing references to recent works regarding Black Carbon, the information on which was unavailable to us at time of writing. Taking into account the Reviewer's comment, the last paragraph of Section 4.1 (page 16795 – beginning line 17) was complemented to read: "Ship-based measurements of black



carbon mass concentration during fall of 1998 in the Barents Sea (Kopeikin et al., 2010) showed that the average values of MBC varied from 160 ng m-3 during northerly and northeasterly winds to 980 ng m-3 when wind blows from the Western Siberia. Most probably, relatively high Mbc values were due to severe fires in boreal zone of the Western Siberia. Based on aircraft measurements in April 1992 over the East Siberian Sea (Hansen et al., 1997), the average values of MBC in the near-ground layer (height of $\sim$100 m) were about 150 ng m-3. Lower Black Carbon concentrations (8-16 ng m-3) for the summer period were reported in works [Sharma et al., 2004; 2013; Stone et al., 2014; Eleftheriadis et al., 2009] on the basis of multiyear (1989-2005) measurements at three Arctic sites: Alert (Canada), Barrow (Alaska), and Zepplin (Svalbard). The average Black Carbon concentrations at these stations are close to our data, obtained over Arctic ocean (region A). It can also be noted that the average Mbc values near the Kola Peninsula (region K) well agree with multiyear (1964-2010) average Black Carbon concentrations (127 ng m-3) at the route-nearest Kevo station in the north of Finland [Dutkiewicz et al., 2014], consistent with the fact that Black Carbon content in the region K is determined by continental sources."

(c) Regression model and its applications are presented on page 16799 (line 17-26) and page 16800 (line 1-9). Dear Reviewer did not explain what should be additionally presented and discussed? 5/6) Sections 5 and 6.

In response to reviewer's comments, Sections 5 and 6 are revised: text, tables, and figures are merged and shortened. In the new text, we tried to define and compare the main specific features of the chemical composition of aerosol and gaseous impurities in the study regions. We also added a comparison with analogous studies in other expeditions. In response to comments of Reviewer #2, we presented a discussion of possible reasons for variations in concentrations of different components in separate periods of expeditions. A difficulty was encountered: (a) to present the characteristics of "typical aerosol composition along the Northern Sea route" and (b) to estimate "the influence of local pollution due to shipping activity and exploration". In this regard, the

following points are in order.

(a) Long-term measurements in each region, with a subsequent statistical processing of a large number of samples are required to determine a typical composition. As we already indicated earlier, individual samples may considerably differ in composition due strong effect of meteorological conditions and synoptic processes. Calculating average over a few samples makes it possible to approach somehow the most characteristic values of the aerosol chemical composition in a particular region (sea) and under specific weather conditions. However, we will not risk stating that precisely typical values are obtained ultimately. In other words, in view of scarcity of data in the study regions, information is still accumulated, and just conservative comments and conclusions are presented.

(b) In the given case, the purpose was to present average characteristics of aerosol chemical composition in separate regions, and not to estimate the influence of local pollution from any sources along the entire route of the expeditions. This investigation will require: accounting for the distribution and current intensity of different aerosol sources on quite a large territory; trajectory analysis of the motion of air masses at different heights, taking into account the particle deposition, etc. It is just unreal to perform a justified analysis in the frameworks of this work. Firstly, we already had to halve the Section 5/6. Secondly, this analysis is of independent interest and is to be published in a separate paper. Nonetheless, in a few cases we added brief comments on possible sources, influencing aerosol chemical composition.

Corrected text of Section 5 (5.1 – 5.4) and updated Tables 6-10 are presented in Supplement pdf-file. In text of Section 5, we made the following changes in figure and table numbering:

(a) old Fig. 12, Fig. 13, and Fig. 16 are removed;

(b) Fig. 14 was renumbered to Fig. 12; and Fig. 15 was renumbered to Fig. 13;



(c) Table 6 was merged with Table 11; Table 7 was merged with Table 12; and Table 8 was merged with Table 13;

(d) changes are introduced in Table 9 and Table 10.

7) Conclusions. In accordance with Reviewer's comment, text of Conclusion section is shortened, namely: – point (a): sentence "The high transparency of the atmosphere over the Arctic Ocean . . ." (page 16817, line 1-3) is removed;

– point (b): the sentence "The boundary layer height showed a similar distribution . . ." (page 16817, line 11-13) is removed;

– point (c): the second paragraph (page 16818, line 4-14) is removed;

– points (e) and (f) are substantially shortened and merged into a single point: (e) Based on results of the chemical analysis of samples, collected in cruises of RV Akademik Fedorov and Professor Khljustin, we determined the average contents of ions and water-soluble elements in aerosol. The sodium ions and chloride ions predominated in aerosol composition throughout the route. Data averaging over three larger water basins (the Arctic Ocean and northern and southern Far East seas) showed that the average concentrations of most ions increase in the southern direction. As a result, the summed concentration of ions, as compared to the Arctic Ocean, increased by 73% over the Bering and Chukchi seas and by a factor of 2.5 at midlatitudes.

The concentrations of water-soluble elements in aerosol composition varied from several tens to several thousandths. Despite certain differences, the concentrations of elements over the Arctic Ocean and Far East seas are grouped in the same way. Members in the first high-concentration group are metals: Zn, Cu, Al, Fe, Ba, Mn, and Sr. Their percentage was 93.7% of the sum of all microelements in the region A and 94.4% over Far East seas. The second group included Ni, Cd, Cr, Se, Pb, and V (5.4 and 5.2% of the total composition). The third group was represented by Mo, Ti, Co, Sb, Li, As,

and Be (0.9 and 0.3% of the total composition). The average percentage of elements in the total sum of soluble components increases in the southern direction from 1.4% in region A to 1.9% in the Japan Sea (except in the Bering Sea, where the average percentage of elements had been 0.8%). With respect to intensity of enrichment of aerosol particles by the elements Zn, Cu, Al, Fe, Mn, Cr, and Cd, the study regions can be lined up in the following order: Okhotsk Sea, Arctic Ocean, Chukchi Sea, Japan Sea, Bering Sea, and East Siberian Sea.

We determined the average content of small gaseous impurities (HCl, HNO3, SO2, NH3) in the atmosphere for separate seas on the Northern Sea Route. From comparison with data of cruises of RV Akademik Fedorov and Professor Khljustin it follows that the concentration of gaseous HCl is several-fold higher, and the concentrations of SO2 and NH3 are several-fold lower, over Far East seas. Analysis of variations in the content of small gaseous impurities and variations in the sum of ions over the Arctic ocean showed the presence of their interrelation with the correlation coeffcient of 0.68. The variations in the SO2 and MSA concentrations over Far East seas are found to be closely interrelated, with correlation coeffcient of 0.77, possibly indicating that DMS was oxidized to give SO2.

The vertical fluxes of water-soluble substances to marine surface are estimated. On the average, deposited ions accounted for about 24.7%, trace elements accounted for 0.4%, and small gaseous impurities accounted for 74.9% of the total amount of substances (Ions+TE+GI), coming from the atmosphere. The maximal fluxes of substances together with ions were observed in water basin of the Chukchi and Bering Seas (28 and 30%), and minimal fluxes were observed over the Japan Sea (16%). The largest contribution (66-83%) to the fluxes of substances was made by the maritime component, i.e., Na and Cl. Among the gaseous species in the Arctic Ocean (region A), chlorine (HCl) accounts for about 16%, nitrogen (NH3+HNO3) accounts for about 22%, and sulfur (SO2) accounts for 62%. Over Far East seas, up to 97 % of fluxes of substances were accounted for by Cl.



8) Reference list is updated to fit revision on the text. So, the reference list now includes new items: Andreas, E.L.: A new Sea Spray Generation Function for Wind Speeds up to 32 m s-1, Journal of Physical Oceanography, 28, 2175–2184, 1998.

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Please also note the supplement to this comment:
http://www.atmos-chem-phys-discuss.net/15/C6611/2015/acpd-15-C6611-2015-supplement.pdf

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Interactive comment on Atmos. Chem. Phys. Discuss., 15, 16775, 2015.

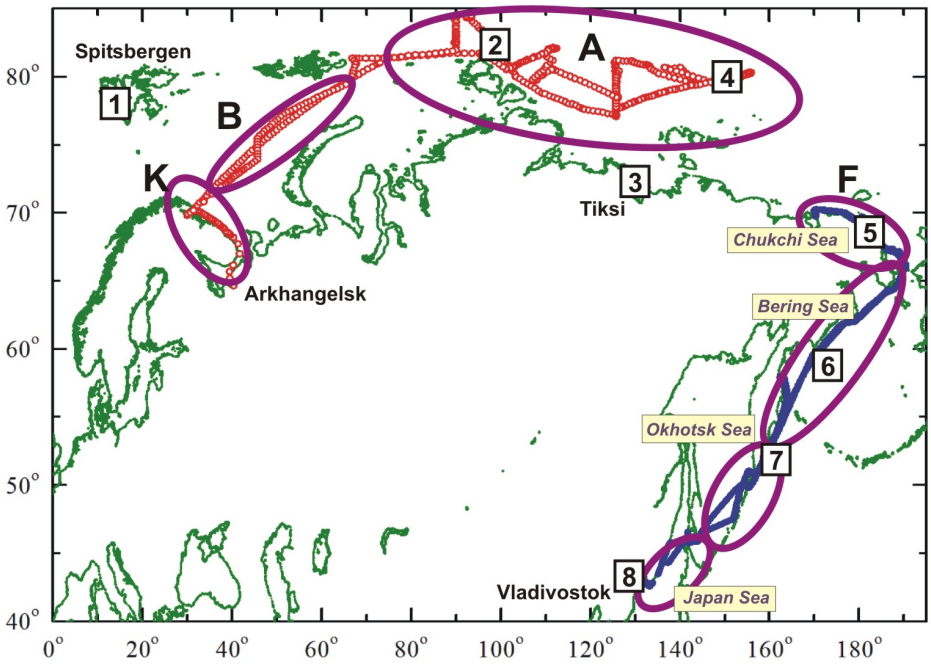


**Fig. 1.** (The updated Fig1.) Map of the routes of Arctic cruises of RV “Akademik Fedorov” and “Professor Khljustin”: numbers indicate the regions where AOD values were calculated; letters and ovals indicate th