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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 # 2

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) Two reviewers provide partly contradictory comments on Section 5/6: on one hand, it is suggested to shorten (merge) two sections; on the other hand, it is suggested to provide an additional interpretation of results. Nonetheless, the text was revised

so as to implement your comment: we added a comparison with analogous results in other expeditions and comments explaining possible reasons for variations in concentrations of different components between separate periods of measurements. It should be noted that chemical composition of aerosol and impurities was difficult to compare adequately for two reasons: (a) for most regions, which we considered, data of other authors are either absent or few; and (b) composition of aerosol samples (even after being averaged over a few samples in a region) is heavily determined by specific features of weather and air transport trajectories. Therefore, the difference in the data between different authors (expeditions) is caused more often by just specific conditions in period of measurements. A characteristic example is the difference in aerosol chemical composition in our data between forward and backward routes of RV Professor Khljustin.

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: 1) old Fig. 12, Fig. 13, and Fig. 16 are removed;

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

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

4) changes are introduced in Table 9 and Table 10.

In addition, the amended list of references. 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.

Clark, P.A.;Fletcher, I.S., Kallend, A.S.;McElroy, W.J., Marsh, A.R.W., Webb, A.H.: Observations of cloud chemistry during longrange transport of power plant plumes, Atmospheric Environment, 18 (9), 1849-1858, 1984.

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snow on Arctic sea ice and in coastal regions, Atmos. Chem. Phys., 4, 2259–2271, doi:10.5194/acp-4-2259-2004, 2004.

Dutkiewicz, V.A., DeJulio A.M., Ahmed T., Laing J., Hopke P.K., Skeie R.B., Viisanen Y., Paatero J., and Husain L.: Forty-seven years of weekly atmospheric black carbon measurements in the Finnish Arctic: Decrease in black carbon with declining emissions, J. Geophys. Res. Atmos., 119, 7667–7683, doi:10.1002/2014JD021790, 2014.

EANET, 2011: Second Periodic Report on the State of Acid Deposition in East Asia. Part II: National Assessments. Niigata, Japan: Acid Deposition Monitoring Network in East Asia (EANET), 305 pp., 2011.

Eleftheriadis, K. Vratolis S., and Nyeki S.: Aerosol black carbon in the European Arctic: Measurements at Zeppelin Station, Ny-Ålesund, Svalbard from 1998-2007, Geophys. Res. Lett., 36 (L02809, 5), doi:10.1029/2008GL035741, 2009.

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Atmospheric Environment, 41, 1604–1615, 2007.

Jung, J., Furutani, H., Uematsu, M., Kim, S., and Yoon, S.: Atmospheric inorganic nitrogen input via dry, wet, and sea fog deposition to the subarctic western North Pacific Ocean, Atmos. Chem. Phys., 13, 411–428, 2013.

Law, K.S., Stohl A., Quinn P.K., Brock C., Burkhart J., Paris J.-D., Ancellet G., Singh H.B., Roiger A., Schlager H., Dibb J., Jacob D.J., Arnold S.R., Pelon J., and Thomas J.L.: Arctic air pollution: New insights from POLARCAT-IPY, Bull. Amer. Meteor. Soc., 95, 1873–1895, doi:10.1175/bams-d-13-00017.1, 2014.

Ohara, T., Akimoto, H., Kurokawa, J., Horii, N., Yamaji, K., Yan, X., and Hayasaka, T.: An Asian emission inventory of anthropogenic emission sources for the period 1980–2020, Atmos. Chem. Phys., 7, 4419–4444, 2007.

Polkin, V.V., Golobokova, L.P.: A comparative analysis of aerosol chemical compositions in complex experiments in Primorye, Atmos. Ocean. Opt., 24(6), 554–565, 2011ÑĄ.

Ponomareva, V.V., Portnyagin, M.V., and Melnikov, D.V.: Composition of tephra from modern (2009-2011) eruptions of the Kamchatka and Kurile islands volcanoes, HERALD Kamchatka regional organization "Scientific training center". Series: Earth sciences. Earth sciences. 20 (2), 23–37, 2012 (in Russian).

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seas, Journal of Geophysical Research, 107 (D24), 4783. doi:10.1029/2001JD001004, 2002.

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Streets, D.G., and Waldhof, S.T.: Present and future emissions of pollutants in China: SO2, NOx and CO, Atmos. Environment, 34, 363–374, 2000.

Sun, J.: Soluble Species in Aerosols Collected on the Route of the First Chinese National Arctic Research Expedition, J. of Glaciology and Geocryology, 24 (6), 744–749, 2002.

Xu, G., Yuan Gao, Y.: Atmospheric trace elements in aerosols observed over the Southern Ocean and coastal East Antarctica, Polar Research, 33, 23973, http://dx.doi.org/10.3402/polar.v33.23973, 2014.

Xu, J., Sun, J., Ren, J., and Qin, D.: Soluble Species in the Aerosols Collected on the Route of the Second Chinese National Arctic Research Expedition. Journal of Glaciology and Geocryology, 27 (2), 205–212, 2005.

Yli-Tuomi, T., Venditte, L., Hopke, P.K., Basunia, M. S., Landsberger, S., Viisanen, Y., and Paatero J.: Composition of the Finnish Arctic aerosol: collection and analysis of historic filter samples, Atmospheric Environment, 37 (17), 2355–2364, 2003.

Zhan, J., Gao, Y., Li, W., Chen, L., Lin, H., and Lin, Q.: Effects of ship emissions on summertime aerosols at Ny–Alesund in the Arctic, Atmospheric Pollution Research, 5, 500–510, 2014.

Zhang, L., Brook, J. R., and Vet, R.: A revised parameterization for gaseous dry deposition in air-quality models, Atmos. Chem. Phys., 3, 2067–2082, 2003.

From the old list of references have been eliminated:

Behera, S.N., Sharma, M., Aneja, V.P., and Balasubramanian, R.: Ammonia in the atmosphere: a review on emission sources, atmospheric chemistry and deposition on terrestrial bodies, Environ. Sci. Pollut. R., 20, 8092–8131, doi:10.1007/s11356-013-2051-9, 2013.

Petrenko, V.V., Ethiridge, D.M., Weiss, R.F., Brook, E.J., Shaefer, H., Severinghaus, J. P., Smith, A. M., Lowe, D., Hua, Q., and Riedel, K.: Methane from the East Siberian Arctic Shelf, Science, 329, 1146–1147, 2010.

Polkin, V.V., Panchenko, M.V., and Golobokova, L.P.: Ion composition of near-water aerosol over White Sea in Augusts of 20032006, Atmos. Ocean. Opt., 20, 911-916, 2007.

Shakhova, N., Semiletov, I., Salyuk, A., Yusupov, V., Kosmach, D., and Gustafsson, Ö.: Extensive methane venting to the atmosphere from sediments of the East Siberian Arctic Shelf, Science, 327, 1246–1250, 2010.

Turekian, K.K.: Oceans, Foundations of Earth Science Series, Prentice Hall, Englewood Clis, USA, 120 pp., 1968.

2) Introduction section and paragraph 2.

We are ready to revise the Introduction section, but Reviewer did not indicate precisely where text is confused. It is also unclear to us what explanations are required in paragraph 2 of Introduction section? Please formulate the comment more clearly. (We note that the Introduction section is revised according to comments of Reviewer #1, and is hoped to be more understandable now).

3) Section 4.1, Paragraph 11:

Taking into account the comments of two reviewers, supplemented by the last paragraph of Section 4.1: "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."

4) Section 4.3, Paragraph 11: In two sentences of this paragraph, we erroneously referred to: Fig. 8e (line 20), Fig. 5c (line 21), and Fig. 5c, d (line 26). It is more correct to write: Fig. 9e (line 20); Fig. 4c (line 21); and Fig. 4c and d (line 26). As to the questions, the explanations will be as follows.

(1 and 2) In this case, we comment (explain) the reasons why small aerosol concentrations are observed at a specific time at a particular location (5 – 10 August, Bering Strait and Chukchi Sea). Because no other measurements in this region are available, we cannot state that the same concentrations will be observed in other cases and, moreover, in other seasons. Nonetheless, small aerosol concentrations over the Bering Strait and Chukchi Sea are not surprising. In our opinion, the aerosol content in these regions is mainly determined by maritime sources. The outflows of continental aerosol from sparsely populated Chukchi Peninsula have minor effect: this region has almost no anthropogenic sources, has permafrost, sparse vegetation, mountainous terrain, and short and rainy summer.

Our AOD measurements on the route around Chukchi Peninsula were unrepresentative. The average AOD values in this region had been: AOD (0.5 μm) = 0.033 on August 4 in the Bering Sea and AOD (0.5 μm) = 0.028 on August 13 in the Chukchi Sea. Thus, the obtained data indicate the small AOD value and are consistent with the text presented.

(3) It is difficult to answer what is the effect of forest fire smokes, because they are characteristic for more southern latitudes, and not for the Arctic Chukchi Peninsula. Seemingly, the far-range transports of smoke aerosol from midlatitudes (boreal zone) to the region of the Chukchi Sea and the Bering Straight can make certain contribution, although no such manifestations were observed in the period of measurements.

Revision in accordance with Reviewer's comments:

Section 1 Paragraph 1. (Define what you mean by "beginning development of the Arctic basin") Reply: Delete the word "beginning" on page 16778, line 4.

Section 1 Paragraph 3: (Re-phrase "own aerosol". Do you mean that local aerosol sources are small?) Reply: Yes, the local (own) aerosol sources in Arctic are weak compared to midlatitudes.

Section 1 Paragraph 4: (Why the focus on methane emissions here? . . .)

Reply: We do not concentrate on methane, but rather present examples of problems, which are urgent in studies of gas composition and indicating that: (1) greenhouse gases from anthropogenic sources increase in content and (2) there is an additional emission due to natural processes. The sentence about methane can be removed; or the entire paragraph can be reformulated more briefly to look: "No less attention was devoted recently to studying the atmospheric trace gas constituents (Duce, 1991; Jacob, 2000; Redington and Derwent, 2002), the content of which grew due to industrial activities (IPCC, 2007, 2013), as well as due to the effect of natural (climatic) factors, e.g., due to methane releases from gas-hydrates in shelf zone, during permafrost melt in subpolar region, and transports of dissolved methane by Siberian rivers (e.g., Sasakawa et al., 2010; Dmitrenko et al., 2011)". In connection with the adjustment of the text in the references are excluded two links:

Section 3.1 Paragraph 7 (Why is the strongest difference in fine AOD noteworthy?)

Replace the last sentence on page 16787, lines 14-16 by the following phrase: Given the general latitudinal decrease of two component of AOD from Arctic to the Japan Sea, the finely dispersed component varies more appreciably (by a factor of 7).

Section 4.1 Paragraph 3 (Why were the highest values observed near the Kola Peninsula?)

Reply: The high values of ozone and BC concentrations are explained by proximity of

vessel to continent, which is the major BC source.

Section 4.1 Paragraph 3 (To me it looks like the enhancement was a factor of 3 (60/20) and not 6?).

Reply: As compared to the region K, Mbc decreased, on the whole, by a factor of 7 (140/20) in the region A. That is, an inaccuracy is made in the text (p. 16793, line 20). It will be more correct to write: ". . .(on the whole, Mbc decreased by the factor of 7 as compared to the region K)."

Section 4.1 Paragraph 6 (Do you mean "fraction" rather than "content"?).

Reply: we mean concentration of particles of coarsely dispersed fraction.

Section 4.1 Paragraph 7 (The Kola Peninsula is not labelled on fig 5. . .).

Reply: The distribution dV/dr in the region near the Kola Peninsula in Fig. 5 is denoted by curve K (as indicated in text above and in Table 4).

Section 4.1 Paragraph 8 (Why are low temperatures important? It is the ice-covered surface . . .).

Reply: In our opinion, both factors are important. Ice coverage of surface reduces generation of maritime aerosol. Low temperatures weaken the photochemical processes of aerosol formation in the atmosphere.

Section 4.1 Paragraph 11 (Why are the values so high in the Kopeikin et al. [2010] study? . . .).

Reply: An additional material is introduced in the last paragraph of Section 4.1, which now indicates how Mbc values increase due to forest fires (see above response to a comment #3 - Section 4.1, Paragraph 11).

Section 4.2 Paragraph 10. (Could you make a second analysis where . . .).

Reply: In response to Reviewer's comments, we made additional calculations after

dataset was processed to remove Ma values at wind speeds larger than 5 m/s. The average latitudinal distribution of Ma for this case is shown in the figure by red line. The obtained results confirm that, when data for large wind speeds are sorted out, the small increase in Ma at high latitudes vanishes and the latitudinal decrease becomes more unambiguous.

(See file Comment_to_Fig8)

Section 4.3 Paragraph 5. (Have previous authors developed such models? . . .)

Reply: There is a number of works devoted to parameterization of aerosol particle concentration as a function of wind speed (Bigg E.K., et al., 1995; Andreas, 1998), in which this dependence is sought in the form lnN=aW+b. In our work, a linear dependence of particle concentration on wind speed (N=aW+b) was used in regression equation; moreover, the regression equation incorporates factors, associated with temperature and precipitation, because these parameters determine the concentration of aerosol particles in the atmosphere. As a result, the quality of approximating the initial data has been better (the determination coefficient turned out to be four times higher when equation (5) was used for data approximation as compared to approximation with the above-mentioned logarithmic dependence). Taking into account the Reviewer's comments, we complemented the first paragraph on page 16800:

1) At the beginning of the paragraph, we added: "Classical equation that relates Na and wind speed looks like log Na = aWS + b (Bigg et al., 1995; Andreas, 1998). In our case of multiple regression, we used a linear dependence of particle concentration on wind speed because this relationship gives better least squares approximation."

2) At the end of paragraph, we added: "First term of (Eq. 5) corresponds to background aerosol concentration under calm conditions and at air temperature of 0C. The second term is associated with wind driven aerosol generation. It is positive, meaning that the greater the wind speed, the larger the aerosol number concentration. The third term is connected with temperature effects on aerosol number concentration. It is negative,

indicating that the greater the temperature, the lower the aerosol number concentration. The last term shows how Na depends on rainfall amount during previous three days. It is negative, implying that the greater the RF amount the lower the Na is. This is due to washing-out effect. It should be noted that regression (Eq. 5) is only valid for data from the route of RV Professor Khljustin. The data, available to us, correspond to limited time frame of about 1 month; therefore, data shortage is the main reason for any future disagreement with experimental data."

3) The reference list is extended to include: 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.

Section 5.1 Paragraph 9. (What do conclude are the sources of the observed aerosol enrichment? . . .)

Reply: These points are explained in the revised Section 5.1.

Section 5.2 (Please provide an interpretation of your results . . ..)

Reply: The Reviewer did not indicate precisely which paragraph needs interpretation. Nonetheless, in the revised Section 5.2 we presented a comparison with data of other authors (e.g., Shevchenko, 2006) and indicated the potential sources of elements in aerosol composition and transport pathways.

Section 5.4 Paragraph 3. (The order of magnitude comment is true for ions but . . .)

Reply: The literature presents many examples in which velocities for gaseous dry deposition are calculated as functions of wind velocity, dry or wet weather, and other factors. In the given case, we did not aim to analyze the influence of different factors on deposition of substances. For the calculations, we chosen maximal velocities of gas deposition on water surface for each gaseous impurity: 2.6 10-2 m s-1 for HNO3, 2.2 10-2 m s-1 for NH3, 2.1 10-2 m s-1 for SO2 (Zhang et al., 2003), and 1.3 10-2 m s-1 for HCl (Clark et al., 1984). In the updated Section 5.4, we also compare our results

with analogous estimates (Shevchenko, 2006), obtained during summer period.

Section 5.4 Paragraph 4. (How do your results compare with previous work?)

Reply: In the updated text, we present a comparison with studies in other regions: at the monitoring station Primorskaya near the Japan Sea (EANET, 2011); in the Atlantic Ocean near Bermuda (Sievering et al., 1989); in the subarctic western North Pacific Ocean (Jung et al., 2013); and in Arctic zone (Shevchenko, 2006).

Section 6.1 Paragraph 5. (Why was the SO42- enrichment smaller over Sea of Japan than . . .?

Reply: The high enrichment of aerosol by sulphates in the Bering Sea is observed episodically and is not characteristic for this region. Most typical coefficients of enrichment of particles by sulphates range from 1.1 to 6.

Section 6.2 Paragraph 6. (Put these results in context of other regions? . . .)

Reply: In the updated text, we note that elevated enrichment of aerosol by certain elements may be observed not only in the Arctic Ocean, but also in Antarctica (Xu et al., 2014), farther removed from anthropogenic sources (see Section 5.2 for more detail).

Section 6.4. Paragraph 4. (Provide some interpretation of the results?)

Reply: In the updated Section 5.2, we present an interpretation of results.

Figure 3: The fonts and line types in Fig. 3 are corrected. The updated Fig. 3 are presented.

Figure 13 when finalizing excluded.

Please also note the supplement to this comment:
http://www.atmos-chem-phys-discuss.net/15/C6630/2015/acpd-15-C6630-2015-supplement.pdf

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

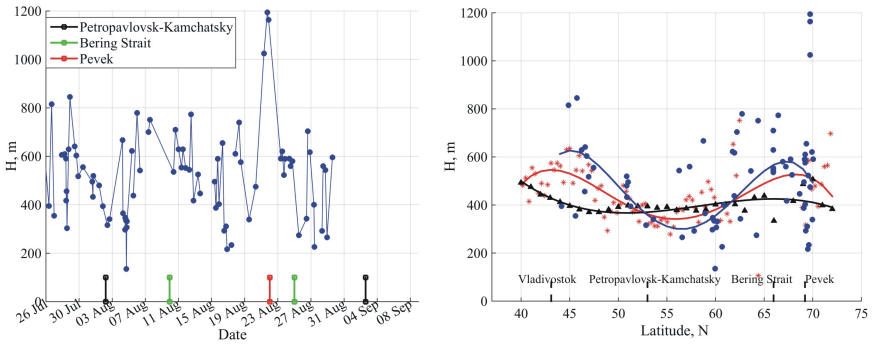
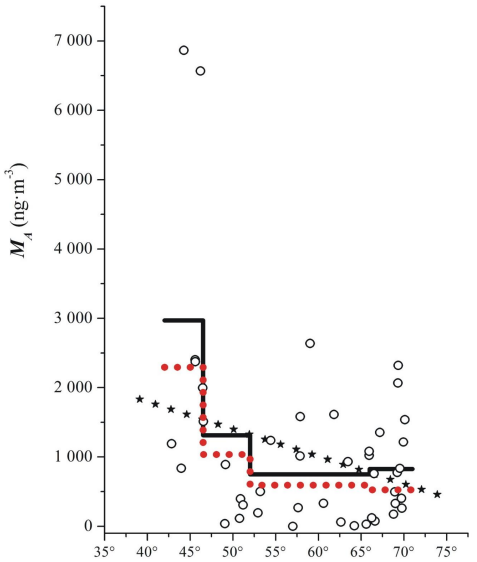


Fig. 1. The updated Fig. 3



Latitudinal distribution of parameters M_A in Far East seas (the red dotted line shows the distribution of wind speeds less than 5 m/s).

Fig. 2. Fig 8*