

Virkkula et al., Aerosol optical properties calculated from size distributions, filter samples and absorption photometer data at Dome C, Antarctica and their relationships between seasonal cycles of sources
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Replies to reviewers, 13 Jan 2022

Dear Editor

We thank the Editor and the reviewers for using their time voluntarily for reading and commenting this long manuscript. Answering the questions and comments hopefully improved the paper. The paper was revised according to most of the suggestions and questions of the reviewers, but not all.

Below the reviewers' texts are written in **bold** font and the replies in standard font.

Major additions to the text are written here an intended paragraph

The largest changes in the paper

1. According to the wishes of Reviewer 2, the data were corrected for inlet and sampling line losses and all results and tables, including those in the supplement. The changes are very small, however, as will be shown below.
2. Description of the loss calculations were added in the supplement.
3. The discussion about the source area analyses using the relative differences was too short. To answer the reviewer's question we added discussion about emissions and transport of BC in the Southern Hemisphere in section 3.4.2, including new references.
4. Both reviewers were wondering whether the residence time of two months is possible. We added discussion about that and new references.
5. The references were crosschecked
6. Added new text has been **highlighted by yellow.**

On behalf of all coauthors,

Aki Virkkula

Detailed replies to Reviewer 1.

Chapter 2.3.1: I assume that your measurements refer to dry aerosol (rh < 40% inside the instruments), correct?

Reply

This is correct. RH was not measured in the Dome C sample air but it can be safely claimed that it was dry. The absolute humidity in the air on the upper plateau is very low and temperature varies from colder than about -20°C in the austral summer down to about -80°C in the austral winter. When air is sampled to the instruments in the measurement containers where temperature is > +10 °C RH decreases to very low values. In the SPO nephelometer RH was recorded and it was mainly ~0% with occasional higher values but also then < 10%.

This is now mentioned in section 2.2.1.

Page 11, lines 7 – 13: To be honest, I do not understand the motivation of this approach. As you mention below, the difference between two consecutive time steps is not purely noise but also includes “true” variability. Maybe you unduly over-estimate the random noise by this procedure?

Reply

This approach was used for getting some estimates of the noises of the scattering and absorption coefficients. Most of the time the real variations were very slow so the noise estimates presented in Table 2 can be considered realistic. Support for this can be obtained from a comparison with the noises presented for the TSI 3wl nephelometer. They are very close to the scattering noises presented in our Table 2.

Page 18, line 9: Please (generally) specify what type of regression is employed (ordinary least square, reduced major axis regression, York regression, ...).

Reply

Ordinary least squares regression was used here. The main purpose of the regression was to study whether there is a statistically significant correlation between the scattering coefficients calculated from the size distributions and the filter samples. This is now mentioned in the text.

Page 25, line 11: Is it realistic, assuming that all particles had a BC core? Is there a convincing reason?

Reply

No, it is not realistic at all to assume that all particles observed at Dome C had a BC core, they definitely don't. Just in case other readers get the same impression the following text was added:

Note that it was not assumed that all particles observed at Dome C had a BC core. The simulations did not use any of the measured number concentrations or absorption coefficients. They were pure simulations with generated size distributions and variable BC core and scattering shell volume fractions. They were conducted to study whether the observed ω_0 vs. feBC(PM₁₀) relationship could be explained with size distributions of coated BC particles. However, considering that the BC particles observed at Dome C most probably have been transported from the surrounding continents for thousands of kilometers it is very likely that at most of the observed BC particles have been coated during aging processes.

Chapter 3.4.2: High RD values over large areas above the southern East Pacific (west of South America) appears somewhat suspicious. From the meteorological point of view, I would have expected such enhanced RD east of South America, i.e. the western part of the South Atlantic.

Reply

We first agreed with this comment and thought it would be some artefact of the method. We found no obvious errors in the calculations, however. There are papers that show that smoke does indeed get transported over the Andes to the Pacific Ocean. The more suspicious region is the western half of South

Atlantic where the RD values are mainly negative. This means that the eBC concentrations observed at Dome C that are associated with air masses from that region belong to the lowest tenth percentile. This is strange because smoke from South America is predominantly transported southeast into the South Atlantic as is discussed in several papers.

The discussion about the source area analyses using the relative differences was very short. Too short. To answer the reviewer's question we added the following discussion about emissions and transport of BC in the Southern Hemisphere in section 3.4.2, including new references. It includes also a more detailed comparison of the footprint difference figure, Fig. 14, with published observations and modeled concentrations on the southern hemisphere.

Smoke from South America is predominantly transported southeast into the South Atlantic (Freitas et al., 2005; Reid et al., 2009; Ulke et al., 2011) and further to the east. African smoke gets first transported westward to the Atlantic Ocean by easterly winds (Reid et al., 2009). Most of this smoke wraps around the the South Atlantic Subtropical High, gets mixed with the South American smoke plume and flows east in the subtropical jet region towards Australia and even further to the remote South Pacific (Singh et al., 2000; Staudt et al., 2002; Chatfield et al., 2002). During the transport to higher latitudes the smoke typically rises to about 5 – 7 km (Singh et al., 2000; Staudt et al., 2002; Reid et al., 2007).

In addition to the eastward transport in the subtropical jet smoke outflow associated with the trade winds, i.e., the dominating easterlies from South America towards the west to the Pacific Ocean has been observed between Equator and 20°S (Freitas et al., 2005, Ulke et al., 2011; Bourgeois et al., 2015). Bourgeois et al. (2015) studied aerosol transport over the Andes from the Amazon Basin to the remote Pacific Ocean by using six years (2007–2012) of data from the Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP) satellite instrument. They found that mainly biomass burning particles emitted during the dry season in the Amazon Basin, are lifted in significant amounts over the Andes. Aerosols reaching the top of the Andes, at altitudes typically between 4 and 5 km, are entrained into the free troposphere (FT) over the southeast Pacific Ocean and they can be traced on average over 4000 km away from the continent indicating an aerosol residence time of 8–9 days in the FT over the Pacific Ocean.

Koch et al. (2009) evaluated BC model predictions of several global models from the AeroCom model intercomparison project. Among several results they presented global maps of modeled BC surface concentrations and annual mean column BC loads predicted by the different models. Fig. 14a displays similarities with both the measurement studies and the global model intercomparison results of Koch et al. (2009). Especially the global maps of annual mean column BC load for GISS sensitivity simulations (Fig. 8 of Koch et al., 2009) have several patterns that are similar to the relative difference RD calculated using Eq. (19) for eBC concentrations measured at Dome C. Here the features of these two figures are compared qualitatively by moving eastwards starting from Africa. The RD in Fig 14a is high above Africa and on the Atlantic Ocean west of Africa, in agreement with the column BC load predicted by all the GISS sensitivity simulations. To the east, between Africa and Australia there is a belt of high RD values, similar to most of the GISS sensitivity simulations. Interesting is that there is even a local minimum over the Indian Ocean in both the RD values and the GISS simulations. The values over Australia are also high in both figures. There is a clear difference east of Australia: the RD values are high even at the meridian of about 160°W – 180°W although the BC load in most of the GISS simulations decreases clearly immediately to the east of Australia. The RD values then decrease to another local minimum at the meridian of about 150°W and then increase towards South America, in agreement with the GISS sensitivity simulations. Over South America the RD values are positive but decrease to negative values on the South Atlantic immediately to the east of the continent. Such a decrease is not in the GISS sensitivity simulations. So the RD values over the western half of the South Atlantic are the most suspicious ones in Fig. 14a. They suggest that the lowest eBC concentrations observed at Dome C were associated with air masses from there. This is not reasonable since the studies cited above show that South American smoke plumes generally flow to the South Atlantic. This discrepancy remains unexplained.

The highest ω_0 are obviously related to sea-spray aerosol (SSA) and biogenic secondary aerosol emissions from the Southern Ocean (Fig. 14b). The differences between the oceanic sectors are not

big but there some: in the Atlantic and Pacific sectors the RD of ω_0 is clearly positive south of latitude 30°S but in the Indian Ocean sector RD is positive south of latitude ~40°S. This can be compared with the Chlorophyll-a (Chl-a) concentrations in five oceanic sectors presented by Becagli et al. (2021). They determined seasonal cycle of Chl-a concentrations in five sectors: Weddell Sea (WS, 60° W–20° E), Indian Ocean (IO, 20–90° E), Western Pacific Ocean (WP, 90–160° E), Ross Sea (RS, 160° E–130° W), and the combined Bellingshausen and Amundsen Seas (BA, 130–60°W). They found that the highest Chl-a concentrations were in the WS, RS and BA sectors and the lowest in the IO and WP sectors. This is in line with the above-described geographical distribution of RD of ω_0 in Fig. 14b suggesting that the oceanic differences are due to biogenic aerosols.

Bourgeois, Q., Ekman, A. M. L., and Krejci, R.: Aerosol transport over the Andes from the Amazon Basin to the remote Pacific Ocean: A multiyear CALIOP assessment, *J. Geophys. Res.*, 120, 8411–8425, <https://doi.org/10.1002/2015JD023254>, 2015.

Chatfield, R., Guo, Z., Sachse, G., Blake, D., and Blake, N.: The subtropical global plume in the Pacific Exploratory Mission Tropics A (PEM-Tropics A), PEM-Tropics B, and the Global Atmospheric Sampling Program (GASP): How tropical emissions affect the remote Pacific, *J. Geophys. Res.*, 107(D16), doi:10.1029/2001JD000497, 2002.

Freitas, S. R., Longo, K. M., Silva Dias, M., Silva Dias, P., Chatfield, R., Prins, E., Artaxo, P., Grell, G., and Recuero, F.: Monitoring the transport of biomass burning emissions in South America, *Environmental Fluid Mechanics*, doi:10.1007/s10652-005-0243-7, 5(1–2), 135–167, 2005.

Koch, D., Schulz, M., Kinne, S., McNaughton, C., Spackman, J. R., Balkanski, Y., Bauer, S., Bernsten, T., Bond, T. C., Boucher, O., Chin, M., Clarke, A., De Luca, N., Dentener, F., Diehl, T., Dubovik, O., Easter, R., Fahey, D. W., Feichter, J., Fillmore, D., Freitag, S., Ghan, S., Ginoux, P., Gong, S., Horowitz, L., Iversen, T., Kirkevåg, A., Klimont, Z., Kondo, Y., Krol, M., Liu, X., Miller, R., Montanaro, V., Moteki, N., Myhre, G., Penner, J. E., Perlwitz, J., Pitari, G., Reddy, S., Sahu, L., Sakamoto, H., Schuster, G., Schwarz, J. P., Seland, Ø., Stier, P., Takegawa, N., Takemura, T., Textor, C., van Aardenne, J. A., and Zhao, Y.: Evaluation of black carbon estimations in global aerosol models, *Atmos. Chem. Phys.*, 9, 9001–9026, <https://doi.org/10.5194/acp-9-9001-2009>, 2009.

Reid, J. S., Hyer, E. J., Prins, E. M., Westphal, D. L., Zhang, J., Wang, J., Christopher, S. A., Curtis, C. A., Schmidt, C. C., Eleuterio, D. P., Richardson, K. A., and Hoffman, J. P.: Global monitoring and forecasting of biomass-burning smoke: Description and lessons from the Fire Locating and Modeling of Burning Emissions (FLAMBE) program, *J. Sel. Topics Appl. Earth Obs. Rem. Sens.*, 2, 144–162, 2009.

Singh, H. B., Viezee, W., Chen, Y., Bradshaw, J., Sandholm, S., Blake, D., Blake, N., Heikes, B., Snow, J., Talbot, R., Browell, E., Gregory, G., Sachse, G., and Vay, S.: Biomass burning influences on the composition of the remote South Pacific troposphere: analysis based on observations from PEM-Tropics-A, *Atmos. Environ.*, 34(4), 635–644, 2000.

Staudt, A. C., Jacob, D. J., Logan, J. A., Bachiochi, D., Krishnamurti, T. N., and Poisson, N.: Global chemical model analysis of biomass burning and lightning influences over the South Pacific in austral spring, *J. Geophys. Res.*, 107, 4200, doi:10.1029/2000JD000296, 2002.

Ulke, A. G., Longo, K. M., and de Freitas, S. R.: Biomass Burning in South America: Transport Patterns and Impacts, in: *Biomass – Detection, Production and Usage*, edited by M. D. Matovic, InTech, Rijeka, Croatia, <https://doi.org/10.5772/19264>, 2011.

Chapter 3.4.4 and Figure 14a: I am particularly amazed about the long residence time of BC emissions in the troposphere before entering continental Antarctica (about 2 months!). Actually this would mean, that BC emissions from any continent of the southern hemisphere would be well stirred before arriving Antarctica. I agree that on average South America is the dominant source region. Nevertheless, this would mean that particular BC concentrations measured in continental Antarctica (derived from atmospheric or ice core data either) would not allow a meaningful source apportionment but merely represent southern hemispheric BC emissions as a whole.

Reply

The residence time can indeed be long if smoke gets high enough. I added the following discussion about residence times including some references to section 3.4.4:

The two-month transport time naturally implies that BC aerosol residence time is also so long. Williams et al. (2002) estimated by modeling that in the upper atmosphere aerosols of 0.065 μm in size have residence times of approximately 1 month and can be transported on a hemispheric scale. Papastefanou (2006) determined residence times of tropospheric aerosols with different methods and obtained a much shorter mean value of 8 days. Kristiansen et al. (2012) estimated accumulation-mode aerosol removal times from the atmosphere using a global measurement data set collected over several months after the accident at the Fukushima Dai-ichi nuclear power plant in March 2011. After correction for radioactive decay, the $^{137}\text{Cs}/^{133}\text{Xe}$ ratios can be assumed to reflect the removal of aerosols by wet and dry deposition. Using this method Kristiansen et al. (2012) obtained removal times for ^{137}Cs of 10.0–13.9 days. The simulations of Croft et al. (2014) with instantaneous injections into layers at 5 km and 7 km yielded mean lifetimes of 14.7 days and 21.1 days, respectively. At higher altitudes removal mechanisms are weaker so lifetimes are longer. These are altitudes where biomass-burning smoke plumes have been observed, as discussed above. Note that these lifetimes, are the e-folding lifetimes τ from $C(t) = C(t_0)\exp(-t/\tau)$. That means, for example, that if biomass burning smoke where BC concentration is 1000 ng m^{-3} – similar to concentrations observed in a Brazilian biomass burning measurement campaign (Brito et al., 2014) – rises to 5 km and gets transported for 60 days the BC concentration decreases to 16.9 ng m^{-3} . With $\tau = 10$ d the concentration would decrease to 2.5 ng m^{-3} . At Dome C the 25th to 75th percentile range of eBC concentration in November is $\sim 2 - 6 \text{ ng m}^{-3}$ (Table S7). As the simple estimation shows this range is consistent with BC emissions of about 1 $\mu\text{g m}^{-3}$ and transport for two months.

The above discussion does not mean that pollution episodes from the surrounding continents could not be transported faster than in two months. For instance, Fiebig et al. (2009) calculated the source areas and transport routes of an elevated aerosol concentration event observed at the Norwegian Troll station in Queen Maud Land. The analysis showed that the origin of the aerosol was biomass burning in Brazil 11 – 12 days before the arrival at Troll. Hara et al. (2010) analyzed several haze events observed at the Japanese station Syowa. They found that the traveling time from Southern Africa and South America to Syowa varied in the range of 4 to 8 days, even faster than to Troll in the episode analyzed by Fiebig et al. (2009). A possible explanation for the considerably longer transport time to Dome C, 2 months, is that it was obtained from the correlation of seasonal cycles of BC emissions and observed concentrations so it is more like a statistical estimate of seasonal transport times. That suggests that on the average the BC emitted in the biomass burning in the surrounding continents circulates the Southern Hemisphere and migrates slowly towards the Antarctic upper plateau.

Croft, B., Pierce, J. R., and Martin, R. V.: Interpreting aerosol lifetimes using the GEOS-Chem model and constraints from radionuclide measurements, *Atmos. Chem. Phys.*, 14, 4313–4325, <https://doi.org/10.5194/acp-14-4313-2014>, 2014.

Kristiansen, N. I., Stohl, A., and Wotawa, G.: Atmospheric removal times of the aerosol-bound radionuclides ^{137}Cs and ^{131}I measured after the Fukushima Dai-ichi nuclear accident – a constraint for air quality and climate models, *Atmos. Chem. Phys.*, 12, 10759–10769, <https://doi.org/10.5194/acp-12-10759-2012>, 2012.

Papastefanou, C.: Residence time of tropospheric aerosols in association with radioactive nuclides, *Appl. Radiat. Isotopes*, 64, 93–100, 2006.

Williams, J., de Reus, M., Krejci, R., Fischer, H., and Ström, J.: Application of the variability-size relationship to atmospheric aerosol studies: estimating aerosol lifetimes and ages, *Atmos. Chem. Phys.*, 2, 133–145, 2002, <http://www.atmos-chem-phys.net/2/133/2002/>.

Figure 15: It is remarkable that concordant for all regions, meridional air mass transport (i.e. transport towards Antarctica) is by far most pronounced between June and October. Is there a link to the polar vortex?

Reply

This was also a good question. Yes, there is indeed a link to the polar vortex. The following text was added to section 3.4.3:

It can be explained by the seasonal cycle of the tropospheric polar vortex. Waugh et al. (2017) described the structures, seasonality, and dynamics of the stratospheric and tropospheric polar vortices and their connections to extreme events at Earth's surface. Estimating from their Fig. 2, the approximate edge of the tropospheric polar vortex of the Southern Hemisphere is at the latitudes of 50°S in January and 30°S in July. This means that in the austral winter the southern parts of South America, Africa and Australia are within the tropospheric polar vortex but not in the austral summer. This is in line with Fig. 15.

Waugh, D. W., Sobel, A. H., and Polvani, L. M.: What is the polar vortex and how does it influence weather, *B. Am. Meteorol. Soc.*, 98, 37–44, <https://doi.org/10.1175/BAMS-D-15-00212.1>, 2017

Technical corrections:

Page 2, line 30: Karpetchko et al. 2005 is absent in the “References”.

Corrected

Page 14, line 25: Stohl et al., 2005 and Pissó et al., 2020 are absent in the “References”.

Corrected

Page 31, line 18: ssp (sigma(sp) is meant, not ssp (sigma(ap).

Corrected

Detailed replies to Reviewer 2.

Major point

In this study, aerosol mass and scattering coefficient were estimated from aerosol number size distributions measured by SMPS and OPC. This approach is common. However, it must be noticed that larger aerosol particles, e.g., coarse particles measured by OPC, can be segregated in the inlet and can be lost efficiently by impaction onto surface/wall of the tubes. Although number concentrations of coarse particles are generally lower than those in sub-micron particles, intensity of scattering light of coarse particles is much greater than that in sub-micron particles. The under-estimation of aerosol number concentrations in coarse modes can lead to mis-estimation of the aerosol properties in this study. Therefore, passing efficiency of aerosol particles into each instrument, particularly OPC, should be taken into account. Actually, authors attempted to discuss difference of scattering coefficient estimated from different procedures in many parts in the manuscript. Furthermore, single scattering albedo estimated in the present study seems to be lower than that in previous works. If aerosol number size distributions were corrected using the passing efficiency, aerosol properties such as aerosol mass, absorption/scattering coefficient, and single scattering albedo are close to true values. I recommend that aerosol number concentrations are corrected using the passing efficiency, and then that the other aerosol properties such as scattering coefficient, mass, absorption coefficient, and single scattering albedo are re-estimated. All descriptions about these points are required to be modified based on results of re-estimations.

Reply

The combined transmission efficiency of the inlet and sample tubing was estimated as a function of wind speed and particle size. The details are presented in the supplement. The transmission-efficiency-corrected number concentrations were then used for recalculating the seasonal variation of the fractions of volume concentration $f_V(\text{DMPS})$ and scattering coefficients $f_{\sigma_{sp}}(\text{DMPS})$ in the size range measured by the DMPS of the respective values calculated from the combined size distributions measured with the DMPS and the OPC at Dome C in December 2007 – July 2009. These fractions were then used for recalculating mass concentrations $m(\text{DMPS}, \text{PM}_{10})$ and scattering coefficients $\sigma_{sp}(\text{DMPS}, \text{PM}_{10})$ and with these all the rest of the results.

It is true that there are clear losses for large particles as can be seen in the supplement Fig S2. However the average wind speed is low, 3.4 ± 1.8 m/s and even when data with $WS < 2$ m/s were rejected due to contamination the average $WS = 3.9 \pm 1.6$ m/s. At this wind speed transmission for $6 \mu\text{m}$ and $10 \mu\text{m}$ particles is $\sim 94\%$ and $\sim 60\%$, respectively but of course lower for higher WS. There was no strong seasonal variation in WS. In addition, the contribution of supermicron particles to number concentration was small and very small in the size ranges where the transmittance losses were significant. If the OPC shows zero particles/cc it is still zero even when it is multiplied with a large number.

As a result the seasonal averages of $f_V(\text{DMPS})$ and scattering coefficients $f_{\sigma_{sp}}(\text{DMPS})$ did not change much. And consequently also the changes to all aerosol optical properties were very small. At the end of this file there is an appendix where the original and transmittance-corrected seasonal cycles of $f_V(\text{DMPS})$ and $f_{\sigma_{sp}}(\text{DMPS})$, aerosol optical properties and mass concentrations are presented.

Just two examples

In Table 4 of the ACPD paper the average $\sigma_{sp}(\text{DMPS}, \text{PM}_{10})$ ($\lambda=530\text{nm}$) = $0.41 \pm 0.88 \text{ Mm}^{-1}$ and in the revised version $0.42 \pm 0.90 \text{ Mm}^{-1}$.

In Table 5 of the ACPD paper the average $m(\text{DMPS}, \text{PM}_{10}) = 171 \pm 255 \text{ ng m}^{-3}$ and in the revised version $173 \pm 260 \text{ ng m}^{-3}$.

There is support for that these values are not drastically wrong. The mass concentration calculated from the PM_{10} filters as the sum of the ion concentrations is $162 \pm 161 \text{ ng m}^{-3}$. Also the percentiles of the mass concentrations $m(\text{DMPS}, \text{PM}_{10})$ and PM_{10} filters are very similar as can be seen both in the original and the revised Table 5 in the appendix.

All results and tables both in the main paper and in the supplement were recalculated. In the figures the changes were so small that they are not visible.

Minor points

Page 6 Line 12-15: Scattering coefficient was estimated using Mie-theory and aerosol number distributions. What is range of scattering angle to integrate light scattering of aerosols? In general, nephelometer does not integrate light scattering aerosols in a whole angle (0-360°).

Reply

The scattering angle θ varies from 0° (forward scattering) to 180° (backscattering). Due to nonideal geometries any real integrating nephelometer indeed does not measure scattering over the whole range. However, here the scattering was calculated for the measured particle number size distributions, there was no need to start simulating the nonidealities of a nephelometer. So the range of the scattering angles was 0° to 180° .

No additions or changes were made to the text.

Page 15 Section 2.7.1: To be honest, I do not follow estimation procedures in the equations (18 and 19), because procedures to obtain the values of "S" were unclear. Additionally, Hirdman et al. (2010) was not listed in the references. Please add short explanation and reference.

Reply

The calculation of the emission sensitivity S was explained at the beginning of section 2.7. It is still there but some new explaining sentences were added. However, it is explained in more detail in the papers cited in the section (Stohl et al., 2005; Hirdman et al., 2010; Pissu et al., 2019).

Page 19 Line 16-20: Scattering coefficient measured using nephelometer can be varied by relative humidity in the nephelometer. Because difference between ambient air temperature (i.e., outside) and temperature in the instrument is larger in the wintertime, sea-salt aerosols can be solid by efflorescence under lower relative humidity conditions. The phase change can modify scattering coefficient. In general, solid particles with irregular shape have larger scattering coefficient than that of droplets. Therefore, this likelihood for the large difference of scattering coefficient in the winter should be discussed.

Reply

Compared with spherical particles, nonspherical particles significantly affect the polarization of light but very little the integrated total scattering of light.

The following text was added:

A hypothetical explanation for the difference between the scattering coefficients at SPO and Dome C could be that in the very dry conditions the particles are not spherical. It is true that the shape of particles affects light scattering. However, it mainly affects the polarization of scattered light: spherical particles do not change the state of the polarization of scattered light but nonspherical particle do. This is used for example in polarization lidars to discriminate ice crystals, dust particles and droplets. However, integral photometric characteristics, such as extinction, scattering and absorption cross sections and single-scattering albedo do not depend significantly on particle shape as is shown in chapter 10 of the textbook by Mishchenko et al. (2002). Therefore nonsphericity is not a likely explanation for the difference.

Mishchenko, M. I., Travis, L., and Lacis, A.: Scattering, Absorption, and Emission of Light by Small Particles, Cambridge University Press, 2002.

Page 24 Line 1: Fig. 11c? (not Fig. 11b?) Please check.

Corrected.

Page 24 Line 14-27: It is true that NPF is important process to supply aerosols into the atmosphere during summer in the Antarctic. But feBC variation is associated with seasonal variations of eBC source strength and transport strength in the Antarctic. Discussion of these issues are needed.

Reply

feBC is the mass fraction of eBC, i.e., it is calculated from $feBC = 100 \times eBC / m$ where m is the total aerosol mass concentration. In the paper the aerosol mass was estimated for two size ranges. But whatever that range is, it is of course clear from the above equation that feBC is affected by both eBC and the total aerosol mass concentration m, not only the eBC concentration.

There is already discussion of the seasonal cycles of BC emissions and transport, they are in section 3.4. We keep that structure of the paper.

Page 26 Line 31 – Page 27 Line 3: Cloud and precipitation are very important for wet deposition of BC. Moreover, diamond dust occurs frequently in the Antarctic plateau. Although this phenomenon is limited lower troposphere, this may contribute greatly to BC deposition onto snow surface. Add short discussion and explanation.

Reply

It is true that diamond dust can act as a removal mechanism. However, in the text mentioned on p.26 line 31 – p.27 line 3 the discussion is based on the removal mechanisms included in the FLEXPART model only. The purpose of these sentences is to try to explain the lines shown in the figure: why is the BC tracer average altitude below that of the inert tracer? What has the model done? So it is not at all tried to offer a full description of BC deposition that could for example be used for comparison with BC sampled from the snow surface.

No changes were done.

Section 3.4.4: In this study, biomass burning is considered as main BC sources in the Antarctic and Southern hemisphere. Basically, I agree with this assumption. Considering economic activity in the countries of the Southern hemisphere, anthropogenic BC emission may be important. Add short discussion and explanation.

Reply

The following text was added:

Stohl and Sodemann (2010) estimated that ship emissions south of 60°S account for half of the total BC concentrations in the lowest 1000 m of the atmosphere south of 70°S in December. However, due to the particle removal processes as described in section 3.4.1 the contribution of ship emissions to BC at Dome C much lower. No quantitative estimations were calculated. Xu et al. (2021) recently presented a new global BC emission inventory for all countries in the world for the years 1960 – 2017. In an Excel file supplement Xu et al. (2021) presents the emissions classified into energy sector, industrial, residential, on-road motor vehicles, other anthropogenic and wildfires. The sums of South American, Southern hemispheric African and Oceanian emissions in the period 2008 – 2013 and the respective contributions of wildfire emissions were calculated. They were $\sim 40 \pm 8 \%$, $\sim 49 \pm 2 \%$ and $\sim 59 \pm 9 \%$ for South American, Southern hemispheric African and Oceanian emissions, respectively. However, only wildfire-emitted BC-particles have the potential to rise high enough to avoid in-cloud or below-cloud scavenging during the transport, as was discussed above.

Xu, H., Ren, Y., Zhang, W., Meng, W., Yun, X., Yu, X., Li, J., Zhang, Y., Shen, G., Ma, J., Li, B., Cheng, H., Wang, X., Wan, Y, and Tao, S.: Updated Global Black Carbon Emissions from 1960 to 2017: Improvements, Trends, and Drivers. *Environmental Science and Technology*, 55 (12), 7869-7879, DOI: 10.1021/acs.est.1c0311, 2021.

Page 29 Line 2-5: Was emission of sea-salt aerosols from sea-ice estimated in this study? If sea-salt aerosol emission was assumed only from open sea-surface, the estimated values are under-estimated strongly in the winter. Previous works presented that sea-salt aerosols were supplied from sea-ice surface (e.g., Frey et al., *Atmos. Chem. Phys.*, 2019; Hara et al., *Environ. Sci. Process Impact*, 2020). More careful discussion and explanation are need to be added.

Reply

The following text was added:

Sea ice and the glacier surfaces especially in the coastal regions are also important sources of sea-salt aerosols (e.g., Frey et al., 2020; Hara et al., 2020). However, the aim was to find a qualitative explanation of the high scattering coefficients observed in winter especially at SPO. The FLEX-SSA offline tool was used for that and it yields a seasonal cycle that at least partially explains the observations. There was no tool for estimating seasonal cycles of sea-salt emissions from sea ice.

Frey, M. M., Norris, S. J., Brooks, I. M., Anderson, P. S., Nishimura, K., Yang, X., Jones, A. E., Nerentorp Mastromonaco, M. G., Jones, D. H., and Wolff, E. W.: First direct observation of sea salt aerosol production from blowing snow above sea ice, *Atmos. Chem. Phys.*, 20, 2549–2578, <https://doi.org/10.5194/acp-20-2549-2020>, 2020.

Hara, K., Osada, K., Yabuki, M., Matoba, S., Hirabayashi, M., Fujita, S., Nakazawa, F., and Yamanouchi, T.: Atmospheric seasalt and halogen cycles in the Antarctic, *Environ. Sci.: Process Impacts*, 22, 2003–2022, <https://doi.org/10.1039/d0em00092b>, 2020.

Page 29 Line 13: What is time zone? UT? or LT?

Reply

It was written on lines 10-11 that t_T was calculated for the prime meridian (0°). There the time of UTC is the same as the local time.

Page 30 Line 2: Is it possible that secondary aerosols were present for one month in the upper troposphere of the Antarctic? More careful discussion is required. If available, add references.

Reply

Yes, it is possible. When aerosols are in the free troposphere the removal mechanisms are very weak. The residence time can be long, as was discussed in the reply for the reviewer 1.

One sentence was added here:

As it was discussed above, Williams et al. (2002) estimated by modeling that in the upper atmosphere aerosol residence time can be ~1 month and Croft et al. (2014) obtained e-folding lifetimes would be ~15 days and ~21 days at 5 km and 7 km altitudes, respectively, so it is reasonable.

Page 30 Line 5-7: Solar radiation shows maximum in December (i.e., summer solstice) in the Antarctic. It should notice that sea-ice extent shows minimum in February – March. In sea ice margin, bioactivity in the ocean is often active, just like blooms. Therefore, approach of sea ice margin with bioactivity (i.e., origins of aerosol precursors) in February – March may relate closely to variations of secondary aerosols such as sulfates and organics. This should be discussed in addition to discussion on NPF and growth.

Reply

This is true. However, we keep the relevant discussion short because all these aspects were discussed in detail in a very recent paper by Becagli et al. (2021), our coauthors who were responsible for the filter sampling and their chemical analyses. It appeared during the review process of the present paper. This text was added here:

Becagli et al. (2021) discussed the relationships between biogenic aerosols (nss SO_4^{2-} and MSA), Chlorophyll-a concentration which is a proxy for phytoplankton, sea ice extent and area, source areas and the transport routes to Dome C and the Southern Annular Mode (SAM). They showed that these relationships are different in early, middle, and late austral summer. Here a subset of the data is analyzed in a method that is complementary to that of Becagli et al. (2021). For more details, see Becagli et al. (2021).

Appendix to the replies to reviewer 2

Changes in the tables when inlet and sample line losses are taken into account.

Table 1. in the ACPD paper.

NO INLET AND SAMPLING LINE LOSSES TAKEN INTO ACCOUNT								
fV(DMPS,PM10), %								
month	fV(DMPS), %		f _{σ_{sp}} (DMPS,467), %		f _{σ_{sp}} (DMPS,530), %		f _{σ_{sp}} (DMPS,660), %	
	average ± std	median	average ± std	median	average ± std	median	average ± std	median
JAN	86.8 ± 10.7	91.6	85.2 ± 9.9	88.7	78.5 ± 12.6	82.5	66.1 ± 15.8	69.6
FEB	85.7 ± 14.0	90.6	82.8 ± 14.3	87.2	76.1 ± 15.9	81.1	64.0 ± 17.5	68.6
MAR	79.0 ± 20.9	88.3	77.0 ± 20.2	85.0	70.2 ± 21.6	78.2	58.6 ± 22.2	65.2
APR	72.8 ± 22.7	83.2	73.6 ± 20.4	80.5	66.4 ± 22.0	72.9	55.0 ± 23.0	59.4
MAY	55.4 ± 24.3	53.1	62.5 ± 20.9	63.9	54.5 ± 22.0	54.6	42.9 ± 22.1	40.7
JUN	49.8 ± 18.3	51.2	61.7 ± 14.6	64.5	52.7 ± 14.6	54.9	39.8 ± 13.2	41.1
JUL	49.5 ± 17.9	50.2	62.2 ± 15.1	65.3	53.4 ± 15.2	55.7	40.8 ± 14.2	41.9
AUG	54.4 ± 15.5	56.0	68.3 ± 12.2	69.9	60.0 ± 13.0	60.7	47.4 ± 13.5	46.7
SEP	62.6 ± 14.6	64.4	73.8 ± 9.8	74.6	66.1 ± 11.4	66.4	54.0 ± 13.3	53.4
OCT	64.6 ± 14.5	66.3	74.1 ± 10.5	74.4	66.4 ± 12.2	66.4	54.2 ± 14.0	53.5
NOV	74.7 ± 13.7	79.1	77.4 ± 10.5	80.2	69.4 ± 12.0	72.0	56.0 ± 13.2	57.7
DEC	80.4 ± 14.4	84.1	80.1 ± 10.9	83.4	72.1 ± 12.1	75.4	57.7 ± 12.4	60.5
year	70.5 ± 21.8	76.7	74.3 ± 17.0	78.4	66.7 ± 18.4	70.3	54.3 ± 19.1	56.2

Table 1 in the revised paper.

INLET AND SAMPLING LINE LOSSES TAKEN INTO ACCOUNT FOR WS > 2 m/s								
fV(DMPS,PM10), %								
month	fV(DMPS), %		f _{σ_{sp}} (DMPS,467), %		f _{σ_{sp}} (DMPS,530), %		f _{σ_{sp}} (DMPS,660), %	
	average ± std	median	average ± std	median	average ± std	median	average ± std	median
JAN	85.7 ± 11.4	91.2	84.2 ± 10.4	88.3	77.3 ± 13.1	81.8	64.6 ± 16.3	68.7
FEB	85.1 ± 14.3	90.5	82.4 ± 14.6	87.1	75.6 ± 16.2	81.0	63.4 ± 17.8	68.4
MAR	78.9 ± 21.2	88.7	76.8 ± 20.7	85.6	70.0 ± 22.1	78.9	58.7 ± 22.7	66.3
APR	72.3 ± 23.5	83.8	72.7 ± 21.7	80.9	65.6 ± 23.2	73.1	54.5 ± 24.0	59.6
MAY	53.8 ± 24.8	48.9	60.5 ± 22.0	61.8	52.6 ± 23.0	51.9	41.4 ± 23.0	37.9
JUN	48.2 ± 18.3	48.7	59.5 ± 15.8	62.7	50.6 ± 15.5	53.0	38.0 ± 13.8	39.4
JUL	48.4 ± 18.3	47.9	60.6 ± 16.0	63.5	52.0 ± 15.9	54.2	39.6 ± 14.7	40.5
AUG	53.5 ± 15.5	55.3	66.3 ± 14.7	69.5	58.1 ± 14.6	60.4	45.7 ± 14.0	46.4
SEP	61.9 ± 14.3	64.1	72.2 ± 13.3	73.8	64.5 ± 13.6	65.9	52.4 ± 14.0	52.8
OCT	63.7 ± 14.2	65.5	72.5 ± 13.2	73.8	64.9 ± 13.7	65.8	52.6 ± 14.2	52.7
NOV	74.0 ± 13.2	77.0	76.9 ± 10.1	79.0	68.8 ± 11.5	70.8	55.2 ± 12.6	56.4
DEC	80.3 ± 13.6	84.0	79.9 ± 10.3	82.9	71.7 ± 11.4	74.9	57.2 ± 11.9	59.8
year	69.2 ± 22.3	73.8	73.0 ± 18.2	77.1	65.4 ± 19.3	68.8	53.1 ± 19.7	54.6

Table 4 in the ACPD paper

	λ (nm)	N	Ave \pm std	Percentiles				
				5	25	50	75	95
Extensive AOPs								
Scattering coefficient, σ_{sp} , Mm^{-1}								
Filter samples								
$\sigma_{sp}(PM_{10})$	550	1765	0.31 \pm 0.31	0.058	0.139	0.24	0.38	0.75
$\sigma_{sp}(PM_{1})$	550	468	0.30 \pm 0.23	0.046	0.118	0.24	0.41	0.75
DMPS-data								
$\sigma_{sp}(DMPS, PM_{0.8})$	467	21987	0.35 \pm 0.64	0.034	0.095	0.19	0.40	1.06
	530	21987	0.27 \pm 0.51	0.026	0.073	0.15	0.30	0.80
	660	21987	0.16 \pm 0.32	0.016	0.044	0.09	0.18	0.46
$\sigma_{sp}(DMPS, PM_{10})$	467	21987	0.48 \pm 0.96	0.052	0.138	0.26	0.52	1.34
	530	21987	0.41 \pm 0.88	0.046	0.120	0.22	0.44	1.12
	660	21987	0.31 \pm 0.73	0.034	0.089	0.16	0.31	0.78
Absorption coefficient, σ_{ap} , Mm^{-1}								
σ_{ap} , calculated using B1999								
$\sigma_{ap}(\sigma_{sp}(DMPS, PM_{10}))$	467	15778	0.015 \pm 0.018	< 0.001	0.0029	0.0096	0.021	0.051
	530	15815	0.013 \pm 0.015	< 0.001	0.0029	0.0085	0.019	0.044
	660	15657	0.012 \pm 0.012	< 0.001	0.0033	0.0077	0.016	0.036
σ_{ap} , calculated using V2010								
$\sigma_{ap}(\sigma_{sp}(DMPS, PM_{10}))$	467	15778	0.016 \pm 0.018	< 0.001	0.0033	0.0099	0.022	0.054
	530	15815	0.012 \pm 0.015	< 0.001	0.0023	0.0076	0.017	0.041
	660	15657	0.009 \pm 0.011	< 0.001	0.0016	0.0057	0.013	0.031
Intensive AOPs								
Single-scattering albedo $\omega_0 = \sigma_{sp}(DMPS, PM_{10}) / (\sigma_{sp}(DMPS, PM_{10}) + \sigma_{ap})$								
σ_{ap} calculated using B1999								
$\omega_0(\sigma_{ap, nsc})$	530	15815	0.93 \pm 0.05	0.84	0.91	0.94	0.97	0.99
$\omega_0(\sigma_{sp}(DMPS, PM_{10}))$	530	15815	0.95 \pm 0.05	0.85	0.92	0.96	0.99	1.00
σ_{ap} calculated using V2010								
$\omega_0(\sigma_{sp}(DMPS, PM_{10}))$	530	15815	0.95 \pm 0.05	0.86	0.93	0.96	0.99	1.00
Scattering Ångström exponent α_{sp} ($\lambda=467/660$ nm)								
$\alpha_{sp}(\sigma_{sp}(DMPS, PM_{0.8}))$	467/660	21987	2.31 \pm 0.28	1.90	2.10	2.25	2.54	2.79
$\alpha_{sp}(\sigma_{sp}(DMPS, PM_{10}))$	467/660	21987	1.35 \pm 0.40	0.77	1.05	1.30	1.66	2.04
Absorption Ångström exponent ($\lambda=467/660$ nm) α_{abs} for $\sigma_{ap} > 3 \delta \sigma_{ap}$								
σ_{ap} calculated using B1999								
$\alpha_{ap}(\sigma_{ap, nsc})$	467/660	15607	0.86 \pm 0.34	0.29	0.67	0.87	1.02	1.37
$\alpha_{ap}(\sigma_{sp}(DMPS, PM_{10}))$	467/660	11475	0.76 \pm 0.47	< 0	0.56	0.82	0.99	1.40
σ_{ap} calculated using V2010								
$\alpha_{ap}(\sigma_{sp}(DMPS, PM_{10}))$	467/660	10270	1.37 \pm 0.38	0.72	1.17	1.37	1.59	2.03

Table 4 in the revised paper, where the DMPS-derived PM₁₀ were estimated by taking inlet and sampling tubing losses into account. The changed values have been highlighted by yellow.

	λ (nm)	N	Ave \pm std	Percentiles				
				5	25	50	75	95
Extensive AOPs								
Scattering coefficient, σ_{sp} , Mm ⁻¹								
Filter samples								
$\sigma_{sp}(PM_{10})$	550	1765	0.31 \pm 0.31	0.058	0.139	0.24	0.38	0.75
$\sigma_{sp}(PM_1)$	550	468	0.30 \pm 0.23	0.046	0.118	0.24	0.41	0.75
DMPS-data								
$\sigma_{sp}(DMPS, PM_{0.8})$	467	21987	0.35 \pm 0.64	0.034	0.095	0.19	0.40	1.06
	530	21987	0.27 \pm 0.51	0.026	0.073	0.15	0.30	0.80
	660	21987	0.16 \pm 0.32	0.016	0.044	0.09	0.18	0.46
$\sigma_{sp}(DMPS, PM_{10})$	467	21987	0.49 \pm 0.98	0.054	0.141	0.27	0.52	1.36
	530	21987	0.42 \pm 0.90	0.047	0.123	0.23	0.44	1.13
	660	21987	0.31 \pm 0.75	0.035	0.091	0.16	0.32	0.80
Absorption coefficient, σ_{ap} , Mm ⁻¹								
σ_{ap} , calculated using B1999								
$\sigma_{ap}(\sigma_{sp}(DMPS, PM_{10}))$	467	15778	0.015 \pm 0.017	< 0.001	0.0028	0.0095	0.021	0.051
	530	15815	0.013 \pm 0.015	< 0.001	0.0028	0.0085	0.019	0.044
	660	15657	0.011 \pm 0.012	< 0.001	0.0032	0.0076	0.016	0.036
σ_{ap} , calculated using V2010								
$\sigma_{ap}(\sigma_{sp}(DMPS, PM_{10}))$	467	15778	0.016 \pm 0.018	< 0.001	0.0032	0.0099	0.022	0.053
	530	15815	0.012 \pm 0.015	< 0.001	0.0022	0.0076	0.017	0.041
	660	15657	0.009 \pm 0.011	< 0.001	0.0016	0.0056	0.013	0.030
Intensive AOPs								
Single-scattering albedo $\omega_0 = \sigma_{sp}(DMPS, PM_{10}) / (\sigma_{sp}(DMPS, PM_{10}) + \sigma_{ap})$								
σ_{ap} calculated using B1999								
$\omega_0(\sigma_{ap, nsc})$	530	15815	0.933 \pm 0.048	0.845	0.909	0.944	0.972	0.985
$\omega_0(\sigma_{sp}(DMPS, PM_{10}))$	530	15815	0.948 \pm 0.049	0.857	0.923	0.959	0.988	1.000
σ_{ap} calculated using V2010								
$\omega_0(\sigma_{sp}(DMPS, PM_{10}))$	530	15815	0.952 \pm 0.047	0.864	0.928	0.963	0.990	1.000
Scattering Ångström exponent α_{sp} ($\lambda=467/660$ nm)								
$\alpha_{sp}(\sigma_{sp}(DMPS, PM_{0.8}))$	467/660	21987	2.31 \pm 0.28	1.90	2.10	2.25	2.54	2.79
$\alpha_{sp}(\sigma_{sp}(DMPS, PM_{10}))$	467/660	21987	1.34 \pm 0.40	0.75	1.03	1.28	1.65	2.03
Absorption Ångström exponent ($\lambda=467/660$ nm) α_{abs} for $\sigma_{ap} > 3 \delta\sigma_{ap}$								
σ_{ap} calculated using B1999								
$\alpha_{ap}(\sigma_{ap, nsc})$	467/660	15607	0.86 \pm 0.34	0.29	0.67	0.87	1.02	1.37
$\alpha_{ap}(\sigma_{sp}(DMPS, PM_{10}))$	467/660	11475	0.76 \pm 0.47	< 0	0.56	0.82	0.99	1.41
σ_{ap} calculated using V2010								
$\alpha_{ap}(\sigma_{sp}(DMPS, PM_{10}))$	467/660	10270	1.38 \pm 0.39	0.73	1.18	1.38	1.60	2.03

Table 5 in the ACPD paper:

	N	Ave \pm std	Percentiles				
			5	25	50	75	95
Mass concentrations							
PM ₁₀ filters, ng m ⁻³	1765	162 \pm 161	30	73	126	201	394
PM ₁ filters, ng m ⁻³	468	82 \pm 64	13	33	66	114	208
m(DMPS, PM _{0.8}), ng m ⁻³	21987	123 \pm 161	12	31	70	150	382
m(DMPS, PM ₁₀), ng m ⁻³		171 \pm 255	23	54	108	199	483
eBC from $\sigma_{ap}(\lambda = 530 \text{ nm})$ calculated using B1999							
eBC(no σ_{sp} correction), ng m ⁻³	15815	2.6 \pm 2.6	0.36	0.94	1.9	3.2	7.6
eBC($\sigma_{sp}(\text{DMPS}, \text{PM}_{10})$), ng m ⁻³	15815	1.7 \pm 2.0	< 0.2	0.37	1.10	2.4	5.7
feBC(m(DMPS, PM _{0.8})), %	15815	2.6 \pm 2.7	< 0.1	0.53	1.62	4.0	7.8
feBC(m(DMPS, PM ₁₀)), %	15815	1.6 \pm 1.7	< 0.1	0.35	1.16	2.5	4.7
eBC from $\sigma_{ap}(\lambda = 530 \text{ nm})$ calculated using V2010							
eBC($\sigma_{sp}(\text{DMPS}, \text{PM}_{10})$), ng m ⁻³	15815	1.6 \pm 1.9	< 0.2	0.29	0.98	2.2	5.3
feBC(m(DMPS, PM _{0.8})), %	15815	2.4 \pm 2.6	< 0.1	0.41	1.48	3.7	7.3
feBC(m(DMPS, PM ₁₀)), %	15815	1.5 \pm 1.6	< 0.1	0.28	1.06	2.3	4.5

Table 5 in the revised paper, where the DMPS-derived PM₁₀ were estimated by taking inlet and sampling tubing losses into account. The changed values have been highlighted by yellow.

	N	Ave \pm std	Percentiles				
			5	25	50	75	95
Mass concentrations							
PM ₁₀ filters, ng m ⁻³	1765	162 \pm 161	30	73	126	201	394
PM ₁ filters, ng m ⁻³	468	82 \pm 64	13	33	66	114	208
m(DMPS, PM _{0.8}), ng m ⁻³	21987	123 \pm 161	12	31	70	150	382
m(DMPS, PM ₁₀), ng m ⁻³	21987	173 \pm 260	23	55	110	200	488
eBC from $\sigma_{ap}(\lambda = 530 \text{ nm})$ calculated using B1999							
eBC(no σ_{sp} correction), ng m ⁻³	15815	2.6 \pm 2.6	0.36	0.94	1.9	3.2	7.6
eBC($\sigma_{sp}(\text{DMPS}, \text{PM}_{10})$), ng m ⁻³	15815	1.7 \pm 2.0	< 0.2	0.36	1.09	2.4	5.7
feBC(m(DMPS, PM _{0.8})), %	15815	2.6 \pm 2.7	< 0.1	0.51	1.60	4.0	7.7
feBC(m(DMPS, PM ₁₀)), %	15815	1.6 \pm 1.7	< 0.1	0.34	1.14	2.4	4.7
eBC from $\sigma_{ap}(\lambda = 530 \text{ nm})$ calculated using V2010							
eBC($\sigma_{sp}(\text{DMPS}, \text{PM}_{10})$), ng m ⁻³	15815	1.6 \pm 1.9	< 0.2	0.28	0.97	2.2	5.3
feBC(m(DMPS, PM _{0.8})), %	15815	2.4 \pm 2.6	< 0.1	0.39	1.47	3.7	7.3
feBC(m(DMPS, PM ₁₀)), %	15815	1.5 \pm 1.6	< 0.1	0.26	1.04	2.2	4.4

