Manuscript ACP-2019-149, Shen et al.: Estimating CCN number concentrations using aerosol optical properties: Role of particle number size distribution and parameterization

Revision and replies to the reviewer's comments, 24-10-2019

We have to thank the reviewer and the editor for their evaluation and their extra voluntary work.

We modified the manuscript to address the reviewer's questions. In the revised text changes are written in red letters. Below the the replies are intended.

Referee #1:

Many thanks for implementing the new Figure 1 - this largely helps to understand the statement on the NCCN-S relationship. However, I still don't agree with the following statement: "The analysis first showed that the dependence of NCCN on supersaturation SS is logarithmic in the range SS < 1.1%.". This infers that finding the ln-relationship is a major discovery. However, there have been several analytical fit functions suggested for the NCCN data over S (Khain et al., 2000; Pinsky et al., 2012; Deng et al., 2013; Pohlker et al., 2016). I wonder why the authors assume that the ln-fit provides advantages over the other functions that have been published before. Moreover, what does "… is logarithmic …" mean? Shouldn't it be rather "… can be described by a logarithmic fit …"? I wonder if the ln-fit has any physical meaning. I suggest that these aspects are discussed in detail in the text. Moreover the aforementioned statement in the abstract probably needs some clarification.

We agree, the statement of the logarithmic dependence on supersaturation was presented too strongly. There is no theoretical reason for the logarithmic dependence, it is simply a fitting to the data.

The statement

"The analysis first showed that the dependence of N_{CCN} on supersaturation SS is logarithmic in the range SS < 1.1%. "

in the abstract was replaced according to the reviewer's suggestion by

"The analysis first showed that the dependence of N_{CCN} on supersaturation SS can be described by a logarithmic fit in the range SS < 1.1%, without any theoretical reasoning.

We thank the reviewer for the new reference list. It revealed that we had not sufficienty read the existing literature. We now read the papers and analyzed the parameterizations presented in them and added related discussion in sections 3.1 and 3.2.

In section 3.1 we added the text below in red letters:

3.1 Site-dependent N_{CCN} - AOP relationships

The averages of AOPs of PM_{10} particles and N_{CCN} at four supersaturations during the analyzed period for each site are presented in Table 2. In general all of them are cleaner than SORPES and more polluted than SMEAR II, based on the average values of σ_{sp}. The average values of N_{CCN} are obviously higher in more polluted air as well as can be seen in the values presented in Table 2. The dependence of N_{CCN} on SS is shown by plotting the averages of the measured N_{CCN} at the six sites at the station-specific supersaturations of the CCN counters (Fig. 1). In all these different types of environments a logarithmic function fits better to the data than the power function

$N_{CCN}(SS) = C\times(SS)^k$. It is not a new observation that the power function is not perfect for describing the N_{CCN} vs. SS relationship. Also other function types have been used in the literature, for instance a product of the power function and the hypergeometric function (Cohard et al., 1998; Pinsky et al., 2012), an exponential function (Ji and Shaw, 1998; Mircea et al., 2005; Deng et al., 2013) and the error function (e.g., Dusek et al., 2003 and 2006b; Pöhlker et al., 2016). In the following analysis of the relationships between N_{CCN}, AOPs and SS we will use logarithmic fittings to the data without any theoretical reasoning.

In section 3.2 we added the text below in red letters. There is an argumentation for using the logarithmic fittings even though there is no theoretical explanation.

The above derivation of the combined parameterization by using the logarithms of SS was fairly straightforward. In the error-function parameterizations of Dusek et al. (2003) and Pöhlker et al. (2016) there are adjustable parameters that affect the argument of the error function. In the parameterization of Ji and Shaw (1998) there is an exponential function where the argument contains the power function of SS and the parameterization of by Cohard et al. (1998) is a product of the power function and the hypergeometric function. If these functions were used for fitting the N_{CCN}(AOP, SS) data it would be would be more complicated to combine the site-dependent parameterizations into a general equation analogous to Eq. (8). The simplicity of the logarithmic fitting makes it most suitable for our approach. The disadvantage of Eq. (8) is that it predicts no upper limit for N_{CCN} at high supersaturations. This is not correct since N_{CCN} cannot be larger than the total particle number concentration and therefore it has to be emphasized that the parameterization presented here is only valid in the range of SS < 1.1%.

In the conclusions we wrote

A logarithmic function was fitted to the N_{CCN} vs. supersaturation SS data in the range SS < 1.1%. For N_{CCN}(AOP) the fitting yielded a logarithmic dependence on SS: $N_{CCN}(AOP) \approx (286 \cdot SAE \cdot \ln(SS/0.093)(BSF - BSF_{min}) + (5.2 \pm 3.3))\sigma_{sp}$.