Elbert, Andreae and Krämer posted a short comment in the interactive discussion of our ACPD manuscript on cloud water chemistry during the HCCT-2010 campaign. We thank the authors for their contribution and would like to reply to it in the following.

We would like to comment on the manuscript published in ACPD by Dominik van Pinxteren et al. (2015) as follows.

1) Concerning page 24315, lines 5-27:

a) Möller et al. (1996) had indeed described the relationship of cloud water total ionic content (TIC) and liquid water content (LWC) by a power law function. In contrast, Elbert et al. (2000) (abbreviated E2000 in the following) confirmed the concept of an inverse relationship, as proposed by Junge (1963), and did not use a power function. To show the inverse relationship between LWC and individual compounds in fog and cloud water (and sometimes the sum of chloride, nitrate, and sulfate) these authors used their own data as well as those published by others. The detailed description of the argumentation and of the computation of the plotted solid curves in their figures (for visualization of the inverse relationship) was presented in section 3.3 of E2000.

E2000 (abbreviation as used by Elbert et al. in their comment) fitted cloud water molalities of the sum of chloride, nitrate, and sulfate as a function of reciprocal LWC for three datasets at two sites (Figs. 1 – 3, section 3.3). Such a reciprocal relationship is a special case of power law functions with a power of -1, because:

$$y = m\frac{1}{x} = mx^{-1}$$

We thus cannot see a contradiction to our statement on P24315 L6-9, which reads:

"Both Möller et al. (1996) and Elbert et al. (2000) concluded from their studies that LWC was the main parameter in controlling cloud water total ionic content (TIC) and that this relationship could be described by a power law function."

b) Not only did Möller et al. acknowledge deviations from their power function, but E2000 also stated that "Since the observed data scatter around an average value, this scattering must be attributed to the various influences which a cloud droplet is subject to, e.g., meteorology, microphysics, gas phase reactions, chemical reactions in the liquid phase and sedimentation (and to some extent to inaccuracies in sample collection, storage and analysis)".

This is correct. However, the scattering is regarded low by E2000, as indicated by statements like "the deviation of the data from the fitted curve […] is relatively small" and "close fit" (both P1111, section 3.1). In addition, E2000 conclude from their survey of literature data that "the amount of soluble material incorporated in cloudwater is fairly invariable at the majority of the sampling sites listed in Table1" (section 3.4). This conclusion is repeated in the summary ("A survey of the available literature leads to the conclusion that, at many locations worldwide, CWL shows surprisingly little variability at a given site") and in the abstract ("A study of published cloudwater data showed that at the majority of the examined locations this product varied little at any given site").

It is these statements we are referring to on P24315 L9-13, which reads:

"While Möller et al. (1996) acknowledged that different air pollution situations lead to strong deviations from the average power function, Elbert et al. (2000) generalized their findings to the statement that at any given site the cloud water loading (CWL, the product of solute concentrations and LWC) would be a fairly constant value (with "fairly constant" being interpreted as max / mean ratio < 5)."

To avoid misunderstandings (and in view of the comment of referee #2 to shorten the introduction) we will omit any reference to the extent of scatter and its respective judgment in the two studies in the revised version and reference only to the broader statement of E2000:

"From a comprehensive literature survey, Elbert et al. (2000) concluded that at any given site the cloud water loading (CWL, the product of solute concentrations and LWC) would be a fairly constant value (with "fairly constant" being interpreted as max / mean ratio < 5)."

c) While van Pinxteren et al. (2015; abbreviated P2015) cite the discussion paper critiquing E2000 by Kasper-Giebl (2002), they unfortunately do not reference the reply to this article by Elbert et al. (2002), which was printed on the pages following Kasper-Giebl's article.

Kasper-Giebl (2002) gave explanations based on the Junge equation, why – conceptually - CWL cannot be a constant. We are referencing these on P23415 L13-16 as follows:

"In a discussion of this proposition, Kasper-Giebl (2002) demonstrated that a constant CWL would imply either constant scavenging efficiencies and substance concentrations in air, or opposite trends of these two parameters, neither of which can be generally regarded as true."

In our view this is an important argument to the discussion. The reply by Elbert et al. (2002) mainly addressed further issues raised by Kasper-Giebl, which we felt would not be relevant to our introductory discussion (e.g. which datasets from the Sonnblick Observatory would adequately follow the proposed inverse relationship and which would not). Related to the referenced statement of Kasper-Giebl (2002), Elbert et al. (2002) replied that "Kasper-Giebl's discussion concerning the relationship between scavenging efficiency and aerosol loading was an interesting contribution, but had not been the subject of E2000". We will explain below why we would disagree here. We agree, however, that a citation of the Elbert et al. (2002) reply would be appropriate and will include it into the revised version of the manuscript.

As a central point of criticism, Kasper-Giebl (2002) expressed the opinion that E2000 had generalized the findings based on the results from their sites to other locations worldwide. To show the limits of this generalization, Kasper-Giebl presented data from the site "Sonnblick Observatory", and stated that (1) a "linear regression between solute concentration and LWC could not be found at that site, and (2) an "average CWL, being relatively constant and characteristic for the site" did not exist. In *their reply, however, Elbert et al. (2002) did not only propose explanations as to why Kasper-Giebl (2002) could not confirm the conclusions reported by E2000, but could also show, using the Mt. Sonnblick data reported by Kasper et al. (1998) and Kasper-Giebl et al. (2000), that the relationship of cloud water concentrations of, e.g., sulfate and LWC could be described simply by an inverse function.*

Reviewing this detailed discussion in Elbert et al. (2002) would be much beyond we can do in our introduction. In our opinion, however, it illustrates the point made by Kasper-Giebl (2002) and referenced in our introduction: The CWL can be “fairly constant” if and only if other conditions such as scavenging efficiency and concentration of the substance in air are “fairly constant” as well (or counteracting their respective influences). This might be the case in some instances (e.g. at sites or during time periods with little variation in CCN concentrations, CCN compositions and size distributions, gas-phase concentrations, etc. etc.), but it cannot be assumed to be generally true.

With regards to an “inverse” or negative relationship of solute molalities/concentrations with LWC in a way of broadly decreasing values with increasing LWC, there is no doubt that it generally exists. The debate – in our understanding – is whether this relationship can generally be described by a simple LWC based function with just some scattering due to minor other influences (which seems to be the notion of E2000) or whether the influences of other parameters on solute concentrations can be similarly or even more important than LWC.

Furthermore, calculation of an average CWL for the sum of the anions chloride, nitrate and sulfate led to a placement of the Mt. Sonnblick site at position #42 (moderately clean environment) in Table 1 in E2000. Finally, Elbert et al. (2002) stated that Kasper-Giebl’s discussion concerning the relationship between scavenging efficiency and aerosol loading was an interesting contribution, but had not been the subject of E2000.

We would disagree here. In our opinion, it was not only interesting, but also relevant to the suggestion of a “fairly constant” CWL at many sites, because it demonstrated (based on the Junge equation) that in order for the CWL to be constant, either the scavenging efficiency and the substance concentration in air would need to be constant or the variation of these parameters would need to cancel out each other’s influences. Neither of this is certainly true.

2) General remarks:

Elbert et al. (2002) included data for very small (as low as 0.01 g m-3) and very large (up to 3.0 g m-3) LWCs (see Figs. 1-5 in Elbert et al. (2002)). The inclusion of such wide LWC ranges allowed the visualization and computation of the inverse relationship. In contrast, P2015 excluded data sets with LWC below 0.15 g m-3 as well as those with more than 0.37 g m-3 (see page 24324, line 5),

These are mean cloud event LWCs. LWCs for individual samples used in the scatter plots of Figure 4 varied between 0.06 and 0.54 g m^{-3}.

and Aleksic and Dukett (2010) (cited by P2015) considered only the sample subset with 0.1≤LWC≤1.0 g m-3.

Which was done by these authors due to doubts about LWC data quality at the very low and high end.

Giulianelli et al. (2014) (also cited by P2015) also excluded data sets with LWC below ~0.1 g m-3 (see their Fig. 7). Because of the omission of the data sets with low and high LWCs it is doubtful that the data of P2015, Giulianelli et al. (2014), and Aleksic and Dukett (2010) can be used to challenge the findings reported by E2000.

The main conclusion of E2000 in our understanding is that in general, solute concentrations are mainly determined by the cloud LWC and therefore the CWL could be expected to be nearly constant at many sites. If this was the case for a wide range of LWCs, then it should be true for smaller LWC subsets as well.

In view of the methodological differences between E2000 and P2015, particularly the data selection by the latter authors, we feel that there is not sufficient basis for the strong statements in P2015, such as "Contrary to some earlier suggestions ...", ...contradicting the earlier conclusions of … Elbert et al. (2000)…", or "...in contrast to earlier suggestions…".

In our understanding, E2000 did not differentiate their conclusions between datasets with extreme ranges of LWC and datasets with smaller ranges. Instead, they propose rather generally, that "our results suggest a universal mechanism governing the composition of cloud droplets" and "that varying amount of condensed liquid water mainly influences the species molalities in cloudwater" (both on P1113) and finally in the summary "that the LWC level is the primary control on the concentration of soluble species in fog and clouds".

What we would like to stress with our results and the cited references is that LWC – while without doubt an important parameter – is not necessarily the primary control on solute concentrations. If the other parameters of the Junge equation, i.e. scavenging efficiency (which is related to microphysical cloud conditions and CCN size distributions and compositions) and substance concentration in the air (both in particles and in the gas-phase) show similar or even higher variances than LWC at a given site, they will strongly impact the observed solute concentrations as well.

We have omitted phrases like the above cited ones in some instances where we felt they could be easily misunderstood, but decided to leave them where the context does justify them in our opinion.

It would be interesting to see what conclusions P2015 and the authors they cite would have reached, had they included data from the full range of LWC.

That is of course not possible to tell with the data available because of the experimental regime of HCCT-2010.

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