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Atmospheric Chemistry and Physics Discussions



# *Interactive comment on* "The climate penalty for clean fossil fuel combustion" *by* W. Junkermann et al.

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General comments

We thank the reviewer for his comments on our paper. Although the reviewer summarizes the contents of the paper the review neglects/oversees the main issue of the manuscript, which is magnitude of the change in particle number emissions due to cleaning of the flue gases and the comparison of the different sources for ultrafine particles. This magnitude of change converts fossil fuel power plant from marginal CCN, but significant sulphur sources into 'marginal' sulphur, but major CCN precursor sources. We will further highlight this in the revised version of our paper.

Reference to previous publications and new data and analysis: Although some of the

material is already published in previous papers we think this is necessary to show the importance and to avoid that the reader has to read several other papers to understand our argumentation. However, the majority of our findings is based on new and up to now unpublished data on plume studies from China and Australia, showing that the process is worldwide and not happening in one location only.

We also included as one of the main points of the paper a previously unpublished comparison of the budget and source strength of several other sources for ultrafine particles that are commonly considered as significant precursor sources for CCN to be able to estimate the importance of the new clean technology emissions.

Contrary to the review statement we do not compare number concentrations of particles between 1 and 10 nm but the sum of particles larger than 10 nm, typically 20 nm and more measured with a standard Butanol-CPC. We show that these particles, assigned in many other publications as CN (condensation nuclei) originate from 1-10 nm directly emitted particles. This size information is crucial for the source appointment as we show in the Junkermann et al Paper 2011.

The manuscript is not intended to discuss in detail the effects inside the clouds. That can be done only with extensive modeling and has been done elsewhere. Here, we are focusing on measurements of an up to now unanticipated source and the comparison of its importance to other well known sources for ultrafine particles purely on experimental evidence. To show this importance we included a description of possible effects. Here we use one of the few available case studies, where we can trace back a change in precipitation to a regional scale doubling of the CCN as an example what can be the impact of a modification of precipitation due to increased numbers of CCN. There are many more publications about these effects, for example Rosenfeld (2000) who found reduced cloud droplet sizes and linked this results to a reduction of precipitation even from one of the sources we measured in 2011, the Port Augusta coal fired power plant. We thus think our impact analysis is thus reasonable and not at all speculative.



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Answers to specific comments

1)The nm sized particles are efficient precursors of CCN. This statement of ours is commented to be not valid by the review. The reviewer states that only a small percentage of these particles (if any) grow to CCN.

It's well known that in a clean maritime atmosphere the majority of CCN are produced from gas phase DMS emissions, over the remote continental boreal forest from new particle production from the gas phase (with a significant contribution from gas phase sulphuric acid, Sipilä et al, 2011). The interaction of these ultrafine particles with atmospheric water vapor producing small droplets is well known since Aitken, (1912) and later Went (1964), though at this time not quantitatively. Nowadays, there are numerous papers available about the growth of the ultrafine particles to CCN, the latest one is (Pierce et al, 2011). Thus, the argument that they are not efficient as precursors may possibly hold in very polluted air with lots of direct emissions, for example in southern China (Qian et al, 2009), but not everywhere else, especially not in pristine remote areas as we state in our manuscript. New, clean, power plant are either planned or already located in remote areas close to the coal as shown in our manuscript (Xilinhot, Inner Mongolia, Kogan Creek, Callide and several others in the Australian outback, etc.)

Growth rate:

The faith of small particles in the atmosphere is either wet or dry deposition or coagulation. As long as they are not significantly coagulating (what we could confirm with our COSMO-ART model), they stay in the atmosphere for several days, slowly growing due to surface reactions until they are finally removed by dry or wet deposition (as CCN). Dry deposition is essential for large particles and for particles close to the ground, However, as we stated in our manuscript the ultrafine particles emitted from the power plants are emitted in an altitude where dry deposition does not play a significant role any more. The duration of the growth to CCN sizes is not critical for our



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analysis. We found about 8 nm per hour in the first two hours of growth, resulting in about six to seven hours to reach CCN size (see below). However, even with half the growth rate or less (Hamed et al, 2010 give a range of 4-8 nm/h), the typical lifetime in the atmosphere of several days is sufficient for the particles to grow into the CCN size range.

The only difference related to slower growth rates or reduced or no growth at all during the night would be a longer transport time. Thus the impact of increased CCN numbers would only appear a few hundred km further downwind and would be even less visible, respectively traceable.

Which fraction of these particles actually grow and act as CCN?

The number of ultrafine particles (CN) emitted is as high, that even a yield of 10 percent for CCN production would be sufficient for a regional scale impact on the average number of CCN. In the Australian experiment (Junkermann et al, 2009) about 15- 20 % of the very fresh excess ultrafine particles were activated, similar to Pierce et al (2011), even larger numbers (about 30%) are currently in discussion. Andreae (2009) reports a ratio of CCN/CN of $0.36 \pm 0.14$.

Model calculations of growth to CCN size

In our COSMO-ART modeling for the 2011 paper (Junkermann et al., 2011) we compared the growth with actual measurements up to 20 nm. COSMO –ART is actually able to calculate the growth to CCN sizes of > 50 nm. We have done these calculations to confirm, that we do not have significant losses due to coagulation and deposition during the subsequent growth. Neither we compared these model results to actual experimental data as we didn't follow the plume that far, nor did we show these model results in the paper as we think, that this is not relevant for the budget considerations. However, we also made such model calculations for the experimental case study in Australia (Junkermann et al, 2009) with a LES model. These modeling results haven't been published up to now, however, a manuscript is in preparation.



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Required size for CCN

The reviewer states that particles of 50 nm are too small to be activated as CCN. Our statement would be true only for remote areas like the remote marine boundary layer. This statement is in contrast to several investigations in Mid-Europe, published for example by Dusek et al, (2006). These authors found in their experiments in Germany that particles down to 40 nm could be activated as CCN. The activation threshold depends on the supersaturation and also slightly on the chemistry. Our sulphuric acid derived hygroscopic particles are active already at the small side of the CCN size spectra at about 50 – 60 nm. This is in also agreement with the recent literature, for example Andreae and Rosenfeld (2008) who reported a threshold of 50 nm for CCN activation.

3)Does flue gas cleaning lead to modified cloud physics?

Here the reviewer argues that the number of CCN with clean air measures decreases and states that 'this is self evident for everybody who has been around in Central Europe in the eighties and nineties'.

Long term measurements of CCN are indeed very scarce. We compare in our manuscript as an example an old dirty power plant which is still in operation (in the US) with smaller new clean power plant (in Germany, China and Australia). The clean ones produce several orders of magnitude higher particle numbers (Junkermann et al. 2011). This result is consistent with the evidence that in central Europe the visibility increased in the eighties and early nineties based on the efforts to clean the air. One of the processes leading to this change is, that the visible (fine and coarse) aerosols were removed in favor of the ultrafine, invisible ones. Now the surfaces for deposition of the ultrafine particles available in the polluted air are removed and the ultrafine particles have a better chance to survive long enough to grow to CCN size. Additionally by catalytic conversion a fraction of the remaining sulphur is directly converted inside the stack to nm sized sulphuric acid droplets. The result is shown in the Hamed et al (2010) paper, also cited in our manuscript. With the reduction of sulphur dioxide emis-



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sions by more than 65% and the frequency of nucleation events by 45%, the number of CCN would be expected to go down also. However, that's not the case even under moderately polluted conditions in mid Europe. Contrary to the expectations the number of CCN/cm3 is stable or even increasing. Hamed et al (2010) are arguing that an additional primary production of sub 100 nm particles is necessary from an unknown source.

CCN, AOD and particle size:

CCN are correlated to AOD as shown by Andreae (2009) (Fig.1). However, this relation is based on the global mixture of co-emission of ultrafine, fine and coarse particles from typical industrial pollution sources and biomass burning. Andreae claims in his paper (2009) that the smaller particles are more efficient CCN and possibly a smaller wavelength for the AOD would give better results. There is also an at least as good correlation shown in the same paper (Fig. 2) with the ultrafine particles, the CN. A ratio of CN to CCN is give as 0.36 (see above), which allows to calculate the number of possible CCN, in case CN measurements are available. Within the recent literature the CN thus are increasingly used as proxies for CCN (Liu et al, 2011). AOD as a proxy can be used from ground based and satellite remote sensing as CCN are typically smaller than the optical remote sensing threshold. A shift of the emission size spectrum, as reported in our manuscript, would affect the first correlation with the AOD but not the correlation with CN.

4)Cloud effects from enhanced CCN, the Ruckstuhl study.

We do not comment in detail in our manuscript on cloud microphysics and radiative effects as this would be a different climate relevant story and, as the reviewer states, requires extensive modeling. That has been done already by several other authors and by ourself (Bangert et al., 2011). We are focusing solely on known precipitation redistribution effects. Naturally both effects are coupled somehow. Cloud microphysics effects can lead either to brighter clouds (or clouds with less transmission) or to lower



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amount of low level stratiform clouds due to faster evaporation of the smaller cloud droplets. The Ruckstuhl study reports a marginal effect of the transmission and a decreasing amount of low level stratiform clouds, which is in prefect agreement with our observations over Western Australia (Junkermann et al, 2009).

The fact, that the Ruckstuhl study did not see any further effect of the shift of the size distribution in the industrial emissions between 1990 and 2010 is obvious. It takes several hours up to a day for the particles to grow to CCN as discussed above. Changes in the emission size spectrum of particles thus can become visible in cloud studies after a transport of about 300 km (assumed 10 m/sec, 8 hours transport in the mid planetary boundary layer) and finally on a larger scale of up to ∼3000 km (5 days). The sites in Switzerland and Germany selected in the study are too close to and partially upwind of the sources. With typical W-E transport patterns in mid Europe we would expect to see effects in Poland or Scandinavia instead.

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