

Interactive comment on “Impact of cloud processes on aerosol particle properties: results from two ATR-42 flights in an extended stratocumulus cloud layer during the EUCAARI campaign (2008)” by S. Crumeyrolle et al.

Anonymous Referee #2

Received and published: 7 March 2012

This paper analyzes measurements of the atmospheric aerosol from one case study in and around cloud in an effort to estimate the net result of cloud processing on the aerosol. It is a potentially interesting case study that may provide a useful look at the processes. Unfortunately, the presentation does not offer a clear picture of the situation nor the processes and there are technical issues to address. As it stands, the paper lacks focus and sufficient technical quality to warrant publication.

Major comments:

C15883

1) A clear and concise discussion of the conceptual model that the authors are basing their analysis on is needed. After some thought, I think I understand what the authors are assuming, but I also do not agree with all aspects of what I believe is their model.

2) More detail in the presentation of the observations is needed as well as a better technical discussion of the limitations of the measurements (e.g. AMS and CVI; see detailed comments). It is important to see the time series of the AMS measurements rather than just a few averages. You need to show how the AMS and SMPS measurements compare across the time series. You suggest that organics increase due to cloud, but your results are neither statistically significant nor properly consider the uncertainties in the measurements.

3) An improved discussion of the processes and impacts is needed. I assume that there were measurements of cloud microphysics made, in which case these need to be included. How do the below-cloud measurements in a particular location compare with the in-cloud measurements? Vertical profiles from below-cloud to above cloud are fundamental to understanding clouds. Cloud microphysical measurements are important if you are to properly discuss the cloud processes and relate them to the measurements downstream of the CVI. Other questions: where is the input aerosol to the cloud; what is the activation point; what is the cycling time of the aerosol through the cloud; how does number change in addition to volume; what are the chemical precursors that may contribute to changes in the size distribution; is collision-coalescence important to the change in the distribution? Cloud-top entrainment is thrown out to explain reductions in aerosol above cloud, but none of the observations address this issue and the attributions that I gather seem extreme.

4) As presented, there is no concept of close vicinity or in the vicinity and hence no relevance. There is no discussion to tie those measurements to the measurements below, in and above the cloud field. The distance scale of the measurements is large (about 500 km) as well as the time scale (2 hours). Your trajectories indicate an increasing continental influence as you move from NW (Newcastle) to SE (Rotterdam).

C15884

Your "Sc vicinity" data are collected towards Rotterdam where the continental influence was strongest. This seems to be the main reason for the strong concentration differences between the 'Sc vicinity' and the cloud region. A transition from continental to marine influence will explain the cloud, the differences in boundary layer depth and the higher concentrations in the cloud-free area. More detail is needed to substantiate your assumption.

Specific comments:

5) Page 33231, lines 14-17 – This sentence did not make sense to me until after I had gone through the paper. But then, what is the importance of this?

6) Page 33231, lines 22-28 – What do you mean by saying that the nitrate and organics "affect" the sulphate and ammonium compounds?

7) Page 33232, line 21 to page 33233, line 6 – You need to be more specific here to help the reader understand your paper. There is a lot known about aerosol-cloud interactions, but you do not convey that. For example: a. It has been long identified that sulphate is produced from the oxidation of S(IV) in clouds. Global models suggest that 50-90% of the atmospheric sulphate is produced in the aqueous-phase. Nitrate is often found in higher concentrations in cloudwater than in the atmospheric aerosol due to the scavenging of HNO₃ or N₂O₅ from the gas phase. Mechanisms for an increase in the organic material (OM) by the aqueous phase have been discussed, but a significant increase in OM in the aqueous-phase relative to the dry particle phase has not been observed in the atmosphere. b. Briefly discuss then how the chemical changes may affect the post-cloud size distribution. c. Then discuss some of the physical scavenging processes, including collision-coalescence. d. Finally, it has been well established that the concentration of cloud-base aerosol particles exerts a significant control on the number concentration of cloud droplets. That process is believed to currently offset climate forcing by greenhouse gases by 10-30% (IPCC, 2007). However, considerable uncertainty remains in how other microphysical and dynamical cloud processes may

C15885

buffer the net impact (Stevens and Feingold, 2009). e. What interaction(s) depends on the type of cloud?

8) Page 33234, line 19 – The brightness temperature, from MODIS, is also shown in Figure 1.

9) Page 33234, line 27 - I don't understand "Nevertheless" in the context presented.

10) Page 33235, lines 6-10 – The cut size and the enhancement factor of the CVI are critical to your estimation of mass concentrations in the cloud. Mountain-top CVIs do not necessarily have the same cut size as airborne CVIs. Our experience is that the Mtn-top CVI has a much lower cut size, and that the airborne CVI cut size estimated from comparison with measurements is higher than the theoretical value. I see no suitable justification here to assume that your cut size was 5 μ m, unless you present comparisons of the residual number concentrations with the details of the cloud droplet size distributions. But you also need to confirm your CVI enhancement factor. Your enhancement factor should be confirmed with CPC measurements from the CVI compared with CDNC from a cloud droplet probe. Were those measurements made?

11) Page 33236, lines 7-22 – The discussion of the PCI could be removed, since you indicate that you have no valid PTOF measurements. More importantly, you talk about the efficiency of the AMS without making the distinction between the transmission efficiency (TE) and the collection efficiency (CE). What you discuss is the TE that is mostly inlet dependent. However, it is also well known that more crystalline particles bounce off the oven before they can evaporate, which is often referred to as the CE (e.g. see paper[s] by Ann Middlebrook at NOAA for a discussion of this issue; pay particular note to the effects of RH). Ammonium sulphate in a relatively dry environment (such as in the CVI sample stream) has one of the lowest values of CE (pure AS in the lab will give a CE of about 25%). Since sulphate is the dominant component of your aerosol throughout this case study, it is important to take into consideration this issue. You can not say that the AMS "quantitatively measures mass loadings" without some other

C15886

corroborating evidence.

12) Page 33237-33238 and Figure 2 – Comments: a. Vertical profiles are critical to understanding clouds and aerosols, and appropriately you conducted many of them (e.g. Figure 2). However, we see no profile data only averages over apparently large times and spaces. Such averages neither allow any resolution of the processes nor do they offer particular value in discussing a net result of the processes because of problems with instrumentation and the lack of a conceptual model. b. Figure 2 is difficult to interpret because of the many colours and some of the explanations. I suggest that you only colour the period when the aircraft was in cloud. c. Your cloud layer was up to about 1km deep, assuming that the “blue” represents cloud (you also indicate about 800 m mean thickness in the text). On the basis of such large cloud depths, the definition of these clouds as SCu does not seem to fit, at least during the morning flt. It looks more like deep stratus or Cu that may have become SCu as the day progressed. A better discussion is needed.

13) Page 33239, Lines 2-9 – Here you state the good things that you are going to assess, but in the end you do not adequately address these issues.

14) Page 33239, lines 11-13 – Better in the discussion of instrumentation where there is also a reference.

15) Page 33240, lines 18-24 – You are telling the reader things that are already known, but not presenting your data in a manner to say anything significant about these processes. The suggestion that entrainment is responsible for the reduced concentrations above cloud relative to the “vicinity” concentrations provides some indication of your overall conceptual model. That is, I assume you are saying the higher concentration aerosol, represented by the ‘vicinity’ aerosol was reduced by the cloud as the aerosol and cloud moved from the NE to the SW. To achieve this, the aerosol in the boundary layer was removed by drizzle and the aerosol was fed from above by entrainment down into the BL via the cloud where it continued to be removed. Perhaps you are right,

C15887

but you would need measurements upwind of your flight track by a hundred kilometres or more to truly say anything about this. For that to have happened you would have needed substantial entrainment at cloud top. Profiles through the cloud will indicate the depth of your transition layer and should help address whether this is reasonable. However, the real problem is that it is unclear whether your in-vicinity measurements have any relationship to the cloud. It is highly unlikely that the concentrations a few hundred kilometres upwind over the North Sea were 8-10 ug/m³, but it would help your case if you can somehow show that is reasonable.

16) Page 33242, lines 14-18 – It is not just about what happened between the morning and afternoon flights. It is about what was going on several hundred kilometres upwind of the flight path on both flights.

17) Page 33242, lines 25-27 – the term “decreasing”, which you use a lot here, implies a steady reduction across some interval. What you show is a step change. However, you have AMS measurements every 23 seconds and you could quite easily demonstrate whether the aerosol is decreasing or simply drops with a time series from the AMS across each flight path. I would not use the measurements from each 23 seconds as this could be noisy due to mass counting statistics. Each 23 second point is a 10 second average, or about 10-15 cm³, and the sampled mass is further reduced due to oven bounce (CE). Using your SMPS distributions, you need to estimate what minimum sampling average of the AMS data is needed to keep the mass sampling noise below about 10%; you need to know your AMS detection limit for each species. Then show a time series of the sulphate, nitrate and organic mass concentrations for each of the two flights. Probably 2-3 minutes averages will suffice.

18) Page 33243, lines 18-22 – You have to work pretty hard to lose 100-300 nm particles in an inlet line, and I doubt that was the problem; here it would help to have a comparison of CVI-CPC and cloud droplet probe measurements. I suspect two main reasons for your high estimated activation diameters, relative to what you expect based on the minimum in your distribution: 1) in the CVI you dry the particles substantially

C15888

and, for a sulphate dominated aerosol, oven bounce may significantly reduce the CE of the AMS; 2) your enhancement factor used in the CVI is incorrect (there is no discussion of this in the instrumental section, but there needs to be).

19) Page 33244, lines 10-18 – A casual reference to entrainment that is another example of the need for a description of your conceptual model. You refer here to sulphate as being produced in cloud and this is recognized as a significant chemical process in the atmosphere, but your results do not indicate an increase in sulphate in cloud. Possible reasons need to be discussed.

20) Page 33245, lines 1-3 – Here again, the absence of a properly explained conceptual model presented early on hurts this paper. Why can the observations not come about without drizzle?

21) Page 33245, lines 11-12 – what are you referring to by the “total N-R PM”?

22) Page 33245, lines 15-16 – If these clouds were truly SCu, then the aerosol would have been cycling between below- and in-cloud with a time constant of perhaps 20-30 minutes (see work by Feingold and Kreidenweis, early 2000) along the wind path. In addition, there would be some limited exchange of aerosol at cloud top and some removal by precipitation. It would be more relevant to compare below-cloud with in-cloud concentrations.

23) Page 33245 – The “sulfate EAP is low”. Again, losses in the CVI line are unlikely to explain this (unless there is something untold). The apparent deficiency in your EAP is more likely based on AMS problems and CVI characterization.

24) Page 33245 – For many years ago, nitrate has often been found in cloudwater at levels comparable to sulphate, in continental situations, and it is more likely to result from the scavenging of HNO₃ (e.g. Leaitch et al., JGR, 1986) or N₂O₅ (e.g. Leaitch et al., JGR, 1988). See also Hayden et al. (JGR, 2008) for results of airborne CVI-AMS measurements of nitrate in cloud.

C15889

25) Page 33246, lines 9-12 – You have many uncertainties in the measurements (your “error” bars are not true uncertainties, just standard deviations) as well as the conceptual model, but you ascribe a 0.12 ug/m³ difference in the organic mass to in-cloud production. Even if I accepted the uncertainty as only described by your SD, the difference between 0.938 and 0.825 is not significant at any reasonable confidence level.

26) Page 33247, line 8 – “framework”.

27) Page 33248, lines 4-7 – the bimodal shape of the below-cloud distribution does not need to rely on drizzle. If these clouds were truly SCu, then the in-cloud and below-cloud air will be mixed to some degree.

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 33229, 2011.

C15890