

We would like to thank the reviewer for taking the time to evaluate our manuscript. In the response below, the reviewer's questions are reproduced in *green* and our replies are shown in black.

1) Throughout the manuscript: I think the authors confuse the term 'reaction rate' and '(reaction) rate constant (or 'rate coefficient'). A reaction rate dc/dt is the product of the rate constant (k) and reactant concentrations (e.g., $[A]$ and $[B]$), i.e. $dc/dt = k [A] [B]$ (cf e.g. Seinfeld and Pandis, 2006) Therefore, a model includes rate constants that are based on laboratory studies. Reaction rates could only be compared between different studies if the same solute concentrations were present. The text should be carefully checked and revised for the correct use of 'reaction rates' and 'rate constants'.

We have now corrected this throughout the manuscript.

2) p. 33850, l. 11: Did any ice form in any of the experiments? If so, might that be differentiated from liquid droplets? Related to this comment: p. 33852, l. 16: Does 'phase transition' only refer here to gas-to-liquid transition or is any ice phase involved?

During the CLOUD8 campaign, ice was formed at the end of the expansions, when the inlet gas lines to the chamber were re-opened. This issue was fixed before CLOUD9. The presence of an ice phase was detected in the Particle Phase Discriminator (PPD-2K) measurements. The onset of ice formation was also seen in the change of SIMONE forward scattering and depolarization signals. The growth of the aerosol droplets always ended well before the ice formation occurred, and no further growth was observed during or after the ice formation.

"Phase transition" refers to liquid/ice.

We have added the following text to the paper, in section 4.3:" During the CLOUD8 experiments at -10°C , the presence of an ice phase was detected from the PPD-2K measurements. The onset of ice formation was also seen in the change of SIMONE forward scattering and depolarization signals. This ice formation was always detected after the pressure decrease had ended, by which time the aerosol growth had also ceased. No further growth of the aerosol was observed during or after the formation of ice. An upgrade in the chamber expansion system between CLOUD8 and CLOUD9 prevented formation of ice during the CLOUD9 experiments."

We added the following text to the instrument description section:

"In addition, the Particle Phase Discriminator mark 2, Karlsruhe edition (PPD-2K) was used to monitor the phase of particles in the size range of 6 to $60\mu\text{m}$, and thus to detect ice formation. Ice particles and water droplets were differentiated by the PPD-2K based on their forward scattering signals (Vochezer et al. 2016).

3) p. 33857, l. 21: Would it be possible to use E-AIM at pressures other than 1 atm? How much difference in the results could be expected if the variable pressure as in the chamber were considered?

Activities and activity coefficients are relatively weak functions of pressure, so for the modest pressure ranges explored in the CLOUD chamber, variations with pressure are not significant.

4) p. 33858/9: Add units to J_c , D_g , J_k in the equations or text before.

The units for D_g are given on page 33858, line 16 (cm^2s^{-1}). The units for J_c are given on page 33858, line 13 (moles per second). We have added the units for J_k (moles per second) to line 2 of page 33859, immediately before equation 3.

5) What are the assumptions of the mass accommodation coefficients for H₂SO₄ and NH₃? What is the limit between 'high' and 'low' accommodation coefficient?

In the paper of McMurry and Grosjean 1985, they consider the limit to be an accommodation coefficient of 6E-6. For gases with lower accommodation coefficients, gas phase diffusion does not play a role, as the uptake is slow (kinetic solution). For gases with higher accommodation coefficients, the uptake on the chamber walls is fast enough that the limiting process is diffusion of the gas to the walls (diffusion limited solution). The rate of diffusion depends also on the coefficient of eddy diffusion, accounting for mixing within the chamber. They consider ammonia to have an accommodation coefficient of the order of 1-5E-8 for their chamber walls, however, in terms of exchange between the gas phase and the chamber walls, the fastest possible rate is that obtained for the diffusion limited approach. As we find that the diffusion limited rate is far too slow to replenish the gas phase, the accommodation coefficient is less relevant, as values less than 1.0 would only lead to an even slower exchange between the walls and the gas phase.

In the CLOUD chamber, during decompression, the rate of diffusion may increase (an increase in the eddy diffusion coefficient), leading to a more rapid exchange between the walls and the gas phase. We have changed the discussion of diffusion in the paper, with the following text replacing the discussion between page 33868, line 6 and line 22:

“The uptake of gases on chamber walls was investigated by McMurry and Grosjean (1985), who pointed out that when the accommodation coefficient of the gas on the chamber walls is high (in their case higher than approximately 6×10^{-6}), transport is diffusion-limited, with the rate of diffusion also depending on the turbulence in the chamber (parameterised by an eddy diffusion coefficient). For their Teflon chamber, they find an accommodation coefficient of $1.2-4.8 \times 10^{-8}$ for ammonia, suggesting that exchange with the walls occurs even slower than the rate of diffusion. For the CLOUD chamber, previous measurements have shown wall loss rates for sulphuric acid of $1.7 \times 10^{-3} \text{ s}^{-1}$ corresponding to a lifetime of approximately 10 min. Assuming ammonia to behave in a similar way to sulphuric acid in the CLOUD chamber, this suggests that the transport between the walls and the gas phase is orders of magnitude too slow to maintain a constant gas phase ammonia mixing ratio during the cloud formation.”

We have also changed the text on page 33869 line 25 to the following:

“It is possible that the pressure change and increased turbulence during the decompression of the chamber lead to a better ventilation of the chamber walls. This would be characterised by a higher eddy diffusion coefficient, increasing the rate of exchange between the gas phase and the walls above the value that is observed during non-decompression periods.”

6) p. 33869, l. 2ff: Is the increased evaporation of ammonia from unactivated aerosol greater because of the larger curvature effect or because of differences in water activity? Does E-AIM take into account both of these effects?

This calculation was only performed accounting for the differences in water content of the aerosol/droplets. EAIM does not account for curvature effects.

7) p. 33869: The section about glyoxal gets a bit lost in this section and should be rather made to a separate section. A few more details should be given in terms of the comparison to NH₃, e.g. how comparable are the solubilities (Henry's law constants) and vapor pressure of these gases? Which parameter determines their uptake/evaporation behavior?

We have put the glyoxal discussion in a separate subsection. We have included information on the effective Henry's law coefficients of glyoxal and ammonia. We would prefer not to speculate on the exact mechanism behind the mobilisation of the glyoxal from the chamber walls at this stage, especially as we have no measurements of ammonia that can be

compared to the glyoxal data. Therefore we did not include a discussion of which parameter determines the uptake or evaporation of the gases. We merely suggest that based on the behaviour of glyoxal, there appears to be a rapid mobilisation of gases from the chamber walls to the gas phase, and this may also be the case for ammonia.

8) *Table 1: It should be added which equilibria K_1 , K_2 , K_3 refer to.* Done

p. 33845, l. 3: CLOUD and CERN should be spelled out here. Done

p. 33858, l. 17: P_{vap} should have lower case p . Done

p. 33864, l. 19: either 'all aerosol is activated' or 'all aerosols are activated': changed to "all aerosol particles are activated"

p. 33867, l. 27/8: 'concentration' misspelled: Fixed

p. 33873, l. 13: Spell out CIGAR: Done

p. 33874, l. 29: received: Fixed

All references end with numbers (Page numbers?). I assume that this happened during copy editing but should be removed. : We shall tell the typesetters to fix this – the numbers are not in the latex file that we are editing, so they must be added by some processing software during typesetting.

Table 3: Is there no T dependence of k_0 ? If so, the minus sign should be removed and '0' added instead.: Correct, there is no temperature dependence. Dash removed, "0" inserted.

Figure 1 (and some other figures) 'shows that' can be omitted: Done.