**Atmospheric
Chemistry
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Discussions**

The Cloud Condensation Nuclei (CCN) properties of 2-methyltetrols and C3–C6 polyols from osmolality and surface tension measurements

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Abstract

A significant fraction of the organic material in aerosols is made of highly soluble compounds such as sugars (mono- and polysaccharides) and polyols, including the 2-methyltetrols, methylerythritol and methyltreitol. The high solubility of these compounds has brought the question of their potentially high CCN efficiency. For the 2-methyltetrols, this would have important implications for cloud formation at global scale because they are thought to be produced by the atmospheric oxidation of isoprene. To investigate this question, the complete Köhler curves for C3–C6 polyols and the 2-methyltetrols have been determined experimentally from osmolality and surface tension measurements. Contrary to what expected, none of these compounds displayed a critical supersaturation lower than those of inorganic salts or organic acids. Their Raoult terms show that this limited CCN efficiency is due to their absence of dissociation in water, this in spite of slight surface-tension effects for the 2-methyltetrols. Thus, compounds such as sugars and polyols would not contribute more to cloud formation in the atmosphere than any other organic compounds studied so far. In particular, the presence of 2-methyltetrols in aerosols would not particularly enhance cloud formation in the atmosphere, contrary to what has been suggested.

1 Introduction

The most important contribution of atmospheric aerosols to Earth's climate, yet still the least understood, is their control of cloud droplet activation and cloud optical properties (aerosol indirect effect) (Forster et al., 2007). While inorganic salts have been shown to be the most efficient materials for cloud activation so far, atmospheric observations have increasingly evidenced the involvement of organic matter in these processes (Novakov and Penner 1993; Liu et al., 1996; Rivera-Carpio et al., 1996; Matsumoto et al., 1997; Ishizaka and Adhikari, 2003; Moshida et al., 2006; Chang et al., 2007). Organic compounds were thus estimated to contribute to up to 63 or 80% of

Cloud Condensation Nuclei (CCN) numbers in marine regions (Novakov and Penner, 1993; Rivera-Carpio et al., 1996; Matsumoto et al., 1997), and 20% at a continental semi-rural site (Chang et al., 2007). The presence of organic compounds was also found to be necessary to account for the CCN numbers in the Amazon basin (Mircea

et al., 2005). A contribution of organic material to CCN could be especially important in pristine environments, such as remote marine regions or the Amazonian wet season, where CCN numbers are limited by the very low aerosol concentrations (e.g. Fitzgerald, 1991; Roberts et al., 2001). Not only cloud formation in these regions is important to understand as a contribution to the global atmosphere, but it also provides

valuable information on the pristine atmosphere, before anthropogenic influence. Over the last decade, a vast number of investigations have attempted to identify organic compounds that might affect cloud droplet activation. A property of these compounds that seems essential in these processes is their solubility in water. Chemical analyses have shown that most aerosols contain a significant fraction of organic compounds of

solubility comparable or even larger than those of inorganic salts (Table 1) such as sugars (mono- and polysaccharides), polyols, and the 2-methyltetrols, methylerythritol and methylthreitol (Claeys et al., 2004; Ion et al., 2005; Kourtchev et al., 2005; Böge et al., 2006). This highly soluble material has been reported to account for up to 5% of the total organic fraction of aerosols in forested (e.g. Graham et al., 2003; Decesari et

al., 2006; Fuzzi et al., 2007), and marine regions (e.g. Simoneit et al., 2004). Polyols and 2-methyltetrols, in particulars, were found in the fine aerosol fraction in forested and rural areas (e.g. Graham et al., 2003; Kourtchev et al., 2005; Böge et al., 2006), making them possible candidates as CCN material in the natural atmosphere. The role of the 2-methyltetrols as CCN material would have tremendous implications for cloud

formation at global scale as these compounds are thought to be produced by the oxidation of isoprene, a globally emitted gas. The CCN efficiencies of saccharides have been previously studied (Rosenørn et al., 2005) and found to be lower than those of organic acids. But the CCN efficiencies of polyols and 2-methyltetrols have not been investigated until now. This work presents the first investigation of the CCN properties

of C3 to C6 polyols and of the tetrols, methylerythritol and methylthreitol.

2 Experimental

The experimental approach used in this work is the one recently developed by Kiss and Hansson (2004) and Varga et al. (2007), and the readers are referred to these
[5] articles for an in-depth description of this method. The principle is to build point by point the complete Köhler curve, $S(r)$, of the compounds of interest by measuring some specific properties of their solutions in water (or salt solutions). The Köhler curve, $S(r)$, describes the supersaturation (or excess water vapor pressure) necessary to activate a particle of radius r into a cloud droplet:

[10]
$$S(r) = \left(a_w \times \exp^{\frac{2\sigma_{sol}M_w}{r\rho_w RT}} \right) - 1,$$
(1)

where a_w is the water activity, σ_{sol} (mN m^{-1}) the surface tension, M_w the molecular weight of water (18 g mol^{-1}), ρ_w the density of water (1 g cm^{-3}), R the gas constant, and T temperature. In this equation, only the parameters a_w and σ_{sol} are related to the compounds studied. All the other parameters are either constant or related to wa-
[15] ter. The values of a_w and σ_{sol} were thus measured experimentally from mixtures of the compounds of interest in water or in salt solutions. To build the whole curve, each mixture was prepared in different concentrations, corresponding to different particle radius, r. The concentrations of organic were varied between 0 and 2 M, and those of salt between 0 and 1 M. The curves were typically built on 5 to 10 points (shown in the Fig-
[20] ures). The particle radius corresponding to the solution concentration was calculated by adding up the volumes of aqueous and of organic materials, the latter assuming the density of the pure organic material. The densities of the organic compounds studied were generally taken from Weast (1970), except for arabitol and the methyltetrols assumed to be 1480 kg m^{-3} and 1460 kg m^{-3}, respectively, by comparison with similar
[25] compounds.

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The surface tension of the solutions, σ_{sol} (mN m^{-1}), was measured with a FTÅ 125 tensiometer, with overall uncertainties of ±2%. The water activity, a_w, was determined from the osmolality of these solutions, C_{osmol} (kg^{-1}), (reduction of water vapor pressure due to the solute), according to:

$$a_w = \frac{\frac{1000}{M_w}}{\frac{1000}{M_w} + C_{osmol}} \quad \text{(Kiss and Hansson, 2004),} \tag{2}$$

where C_{osmol} was measured experimentally with a KNAUER K-7000 vapor pressure osmometer. This method has been shown to provide a_w with an excellent accuracy compared to literature data (Kiss and Hansson, 2004), and less than 2% of errors for up to 1.5 mol kg^{-1} of solute. Uncertainties on these measurements were between ±4% (intermediate concentrations of organics) to ±12% for very dilute and very high concentrations of organics.

The uncertainties on C_{osmol} and σ_{sol} resulted in uncertainties between ±4% and ±7% on $S(r)$. The critical supersaturations, S_c, had the lowest uncertainties, ±4%, because they corresponded to intermediate organic concentrations, where the uncertainties on C_{osmol} were minimal. Note that this method employs the original Köhler Eq. (1), and therefore avoids altogether the uncertainties contained in the simplified equation and in the Van't Hoff factors.

A first series of experiments focused on determining the Köhler curves for the pure organic compounds, glycerol (C3), erythritol (C4), arabitol (C5), mannitol (C6), their di-acid analogs, malonic acid (C3), succinic acid (C4), adipic acid (C6), and the two 2-methyltetrols, from their solutions in water. The Köhler curves presented in this work were determined for a dry particle diameter of 60 nm. Because organic material is always accompanied with inorganic salts in aerosols, which can dramatically affects their Köhler curves (Bilde and Svenningsson, 2004), a second series of experiments determined the Köhler curves for the organic compounds mixed with sodium chloride and ammonium sulfate. All the solutions used for these measurements had a composition of 20 weight percent salt relative to organic weight.

2.1 Chemicals

2-methylerythritol and 2-methylthreitol were custom synthesized by InnoChemie GmbH, Germany. Briefly, the synthesis proceeded first to Compound 1 (Fig. 1), which was isolated in >98% purity. After hydrolysis, benzaldehyde 2 was removed by re-
5 peated azeotropic distillation with water to furnish 3 in nearly quantitative yield contain-ing ∼10% of water. Residual water was removed by repeated azeotropic distillation using ethanol. The final product was dried under reduced pressure in order to remove ethanol. The estimated purity of 2-methylerythritol and 2-methylthreitol was >95%.

All other compounds were commercially available from the manufacturers: Malonic
10 acid (Aldrich, 99%), succinic acid (Aldrich, ≥99%), adipic acid (Aldrich, 99%), glyc-erol (Aldrich, ≥99.5%), erythritol (Aldrich, ≥99%), arabitol (Aldrich, ≥99%), mannitol (Aldrich, 98%), sodium chloride (Aldrich, ≥99%), ammonium sulfate (Aldrich, ≥99.5%).

3 Results and discussion

3.1 Organic/water mixtures

15 The measurements of C_{osmol} and σ_{sol} made in this work are summarized in Table 2. Their variations with the organic concentration, $c(M)$, are represented by their best fit to empirical first- to third-order expressions. Note that these non-linear variations were larger than the uncertainties and reproducible between water and salt solutions, indicating that they were real and not experimental artifacts.

20 The Köhler curves for the polyols and di-acids are shown in Fig. 2, and for the 2-methyltetrols, in Fig. 3. Previous measurements of the CCN properties for malonic acid with the same experimental method reported an excellent agreement with litera-ture data (Varga et al., 2007). The curves obtained in this work were also in excellent agreement with previous measurements for malonic (Prenni et al., 2001; Giebl et al.,
25 2002; Hori et al., 2003), succinic (Corrigan and Novakov, 1998; Hori et al., 2003; Bilde

and Svenningson, 2004), and adipic acid (Cruz and Pandis, 1997; Corrigan and No-vakov, 1998; Prenni et al., 2001; Hori et al., 2003; Bilde and Svenningson, 2004). This good agreement with techniques as different as HTDMA (e.g. Prenni et al., 2001) and CCN counters (e.g. Corrigan and Novakov 1998; Giebl et al., 2002; Bilde and Sven-ningson, 2004) demonstrates the validity of the technique used in this work, even when applied to compounds having significant surface-tension effects, such as succinic acid.

The Köhler curves obtained for the polyols (Sc=0.52–0.63±0.02%) and the 2-methyltetrols (Sc=0.57–0.68±0.02%) showed that the critical supersaturations of these compounds were all higher than those of their analogue di-acids (Sc=0.44–0.52%) (Figs. 2 and 3). This demonstrates that, in contrast to what ex-pected, a high solubility is not necessarily equivalent to a high CCN efficiency. These results are generally in line with the low CCN efficiencies previously measured for other highly soluble compounds, mono- and di-saccharides (Sc=0.55–0.85%) (Rosenørn et al., 2005). Examining the Raoult terms for the compounds studied in this work pro-vides the explanation for their limited CCN efficiencies: while the high CCN efficiencies of sodium chloride and ammonium sulfate result from their large Raoult term, and cor-respondingly high osmolalities, the osmolalities of the polyols and methyltetrols were comparably low (Table 2). For instance, for c=0.1 M the expressions in Table 2 give:

C_{osmol}=210×10^{-3} kg^{-1} for (NH$_4$)$_2$SO$_4$,

=174×10^{-3} kg^{-1} for NaCl,

=110 to 117×10^{-3} kg^{-1} for the organic acids,

=97 to 112×10^{-3} kg^{-1} for the linear polyols, and

=61 and 67×10^{-3} kg^{-1} for methylerythritol and methylthreitol, respectively,

(all with uncertainties of ±14×10^{-3} kg^{-1}). Raoult's law implies that osmolality is pro-portional to the concentration of solute in these solutions. Thus, it appears that the differences between the osmolality values of different classes of compounds reflect the degree of dissociation of these compounds: the polyols, that do not dissociate, pro-duce only one molecule of solute per dissolved molecule and their osmolalities are low.

Organic acids partly dissociate, producing between 1 and 2 molecules of solute, and their osmolalities are roughly 1.5 times those of the polyols. Inorganic salts dissolve completely, producing 2 (NaCl) or more (($(NH_4)_2SO_4$)) molecules of solute, and their osmolalities are 2–3 times larger than those of the polyols. Differences between the osmolalities of individual polyols suggest that other factors than the degree of dissociation affect the Raoult term, but to a smaller extent.

Surface tension effects would partly compensate for low water activities and improve the CCN efficiencies, as it has been shown for organic acids (Facchini et al., 1999). The surface tensions measured in this work as function of the molar concentration, $c(M)$, are summarized in Table 2. For c=0.1 M, the surface tension for solutions of adipic and succinic acid were σ_{sol} (0.1 M)=66 and 70 (±1) mN m^{-1}, respectively. None of the linear polyols displayed any significant surface tension effect (σ_{sol} (0.1 M)~71±1 mN m^{-1}), but the 2-methyltetrols displayed a small effect: σ_{sol} (0.1 M)~70 mN m^{-1} for both of them. These effects contributed to lower their critical supersaturation, but not enough to be better CCN than the inorganic salts or even the organic acids.

3.2 Organic/salt/water mixtures

The measurements of C_{osmol} and σ_{sol} for the organic/salt/water mixtures are also presented in Table 2 and the Köhler curves in Figs. 4 and 5. For adipic acid with sodium chloride, our results are in agreement with those of Bilde and Svenningsson (2004) showing a strong reduction the critical supersaturation compared to the water mixtures (Sc=0.52% in water and 0.42% in NaCl, ±0.02%), and a slight enlargement in the critical diameter. This agreement shows that our experimental method remains valid when applied to organic/salt mixtures.

Ammonium sulfate was found to have less impact on the critical supersaturation than sodium chloride (Sc=0.51%). This difference is likely to result from the different pH of these salts: sodium chloride solutions are slightly basic (pH=7–8) and favor the dissociation of weak acids, while ammonium sulfate solutions are slightly acidic

(pH=5.5–7) and limit their dissociation.

For mannitol, the critical supersaturation was reduced by both salts: from Sc=0.62% in water, to 0.45% in NaCl, and 0.54% in $(NH_4)_2SO_4$. This suggests that mannitol is only partly soluble in water, in agreement with the moderate solubility reported in Table 1. As with adipic acid, the critical supersaturation was less reduced by ammonium sulfate than by sodium chloride. By contrast, the critical supersaturation of methylthreitol was hardly affected by the presence of either salt: Sc=0.69% in water, and 0.66% NaCl, and 0.68% in $(NH_4)_2SO_4$. This lack of effect of salt suggests a very large solubility of this compound in water, in line with the solubility of threitol (Table 1). Interestingly, the critical supersaturation for methylerythritol was increased by both salts: Sc=0.58% in water, 0.60% in NaCl, and 0.69% in $(NH_4)_2SO_4$. A possible explanation for this surprising result is that this compound, as erythritol (Table 1), is only partly soluble in water. However, unlike the di-acids and polyols, the non-soluble part would be liquid not solid, and might form a film at the surface of the droplets, which would limit the uptake of water and therefore the CCN efficiency.

4 Conclusion and atmospheric implications

In this work, complete Köhler curves for a series of C3–C6 polyols and methyltetrols were determined experimentally by measuring the osmolality and surface tension of their organic/water and organic/salt/water solutions. The excellent agreement of the Köhler curves obtained for malonic, succinic, and adipic acid with literature data obtained with other techniques demonstrates the validity of this experimental method, even when applied to compounds having significant surface-tension effects. The Köhler curves for the C3–C6 polyols and the 2-methyltetrols show a slightly lower CCN efficiency than organic acids, both in water and in the presence of salts. These results demonstrate that a high water solubility is not necessarily equivalent to a high CCN efficiency. They are also in line with the low CCN efficiencies determined previously for saccharides. Thus, sugars and polyols would not contribute more to cloud formation

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in the atmosphere than any other organic compounds studied so far. In particular, the presence of 2-methyltetrols in aerosols would not particularly enhance cloud formation in the atmosphere, contrary to what has been recently suggested (e.g. Silva Santos et al., 2006; Meskhidze and Nenes, 2006).

5   However, under certain conditions, it is possible that this highly soluble material contributes to activate smaller CCN. In pristine environments that are limited in CCN numbers, such as remote marine regions and the Amazonian wet season, this might somewhat increase the CCN numbers and, in turn, affect droplet size. The importance of such effect remains however to be determined.

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Table 1. Solubility in water for the compounds discussed in this work.

Compound	Solubility [$g\,L^{-1}$]	Reference[a]
Polyols:		
Methyl threitol	8800[b]	
Methyl erythritol	637[c]	
Glycerol	Inf.	Saxena and Hildemann, 1996
Threitol	8800	Cohen, 1993
Erythritol	637	Cohen, 1993
Arabitol	1510–2110	Saxena and Hildemann, 1996
Mannitol	216	Hu, 1998
Saccharides:		
Fructose	4074	Washburn, 1927
Mannose	2500	Windholtz, 1983
Sucrose	2000	Windholtz, 1983
Glucose	909	Windholtz, 1983
Lactose	200	Windholtz, 1983
Maltose	93	Washburn, 1927
Di-acids:		
Malonic acid	1610	Saxena and Hildemann, 1996
Succinic acid	88	Saxena and Hildemann, 1996
Adipic acid	25	Saxena and Hildemann, 1996
Inorganic salts:		
Ammonium Sulphate	706	Weast, 1970
Sodium Chloride	357	Weast, 1970

[a]Complete references in auxiliary material
[b]Assumed identical to threitol
[c]Assumed identical to erythritol

The CCN properties of 2-methyltetrols and C3–C6 polyols

S. Ekström et al.

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Table 2. Variations of the surface tension and osmolality with the concentration of organic compound, $c(M)$. In brackets: number of measurements.

| Compound	Surface tension (mN m^{-1})	r^2	Osmolality ($\times 10^{-3}$ kg^{-1})	r^2
		Water		
Glycerol	$-1.8c+71.6$ (6)	0.97	$-121.8c^2+718.7c+41.4$ (6)	1.00
Erythritol	$-1.8c^2-0.6c+71.8$ (6)	0.91	$1107.9c-14.0$ (6)	1.00
Arabitol	$-16.3c^2-3.1c+71.2$ (8)	0.76	$1018.5c+3.2$ (8)	1.00
Mannitol	$1.4c^2-4.6c+71.9$ (6)	0.94	$1072.4c-9.3$ (6)	1.00
Methylerythritol	$4.7c^2-22.4c+71.9$ (12)	1.00	$-310.6c^3+936.8c^2+249.9c+32.9$ (12)	1.00
Methylthreitol	$-6.7c^2-8.1c+71.2$ (7)	0.97	$312.1c^2+236.5c+34.4$ (7)	0.99
Malonic acid	$1.6c^2-7.8c+71.5$ (6)	0.98	$978.6c+19.5$ (6)	1.00
Succinic acid	$-19.4c+72.1$ (6)	0.95	$1018.1c+9.8$ (6)	1.00
Adipic acid	$193.1c^2-72.3c+71.0$ (6)	0.98	$1016c+8.3$ (6)	1.00
Sodium Chloride	$-2.4c+71.3$ (8)	0.62	$1800c-6.5$ (8)	1.00
Ammonium sulfate	$-2.7c+70.8$ (9)	0.93	$1946c+15.3$ (9)	1.00
		Ammonium Sulphate		
Mannitol	$3.3c^2-7.4c+72.1$ (9)	0.97	$1322.1c-18.4$ (9)	1.00
Methylerythritol	$-0.4c^3+0.7c^2-15.3c+73.4$ (9)	1.00	$500.8c^2+201.3c+32.8$ (9)	1.00
Methylthreitol	$4.2c^3-10.3c^2-8.3c+72.7$ (9)	1.00	$442.3c^2+349.3c+20.0$ (9)	1.00
Adipic acid	$-4189c^3+981.1c^2-105.2c+72.2$ (6)	1.00	$-19\,596c^3+6291c^2+425.4c+25.8$ (6)	1.00
		Sodium Chloride		
Mannitol	$-2.7c+70.9$ (9)	0.95	$1655.7c-19.7$ (9)	1.00
Methylerythritol	$12.8c^3-29.9c^2+3.9c+71.4$ (9)	1.00	$528.1c^2+338.2c+32.1$ (9)	1.00
Methylthreitol	$-4.3c^2-7.2c+72.7$ (9)	0.99	$449.6c^2+351.7c+9.0$ (9)	1.00
Adipic acid	$-33.2c^3+688.4c^2-62.3c+69.8$ (6)	1.00	$15\,425c^3-31\,159c^2+2577c+21.5$ (6)	0.99
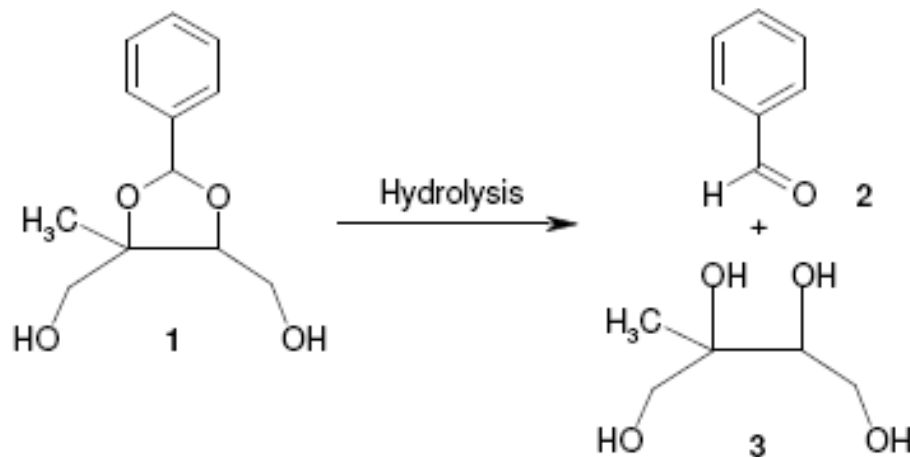
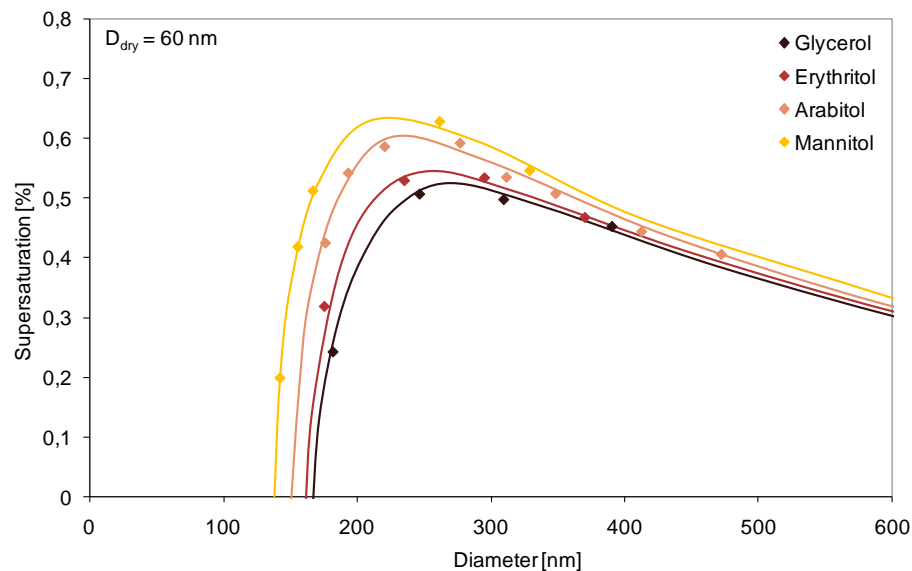**Fig. 1.** Details of the molecular structures of the intermediates in the synthesis of the methyl-tetrols (courtesy of Innochemie GmbH).

Fig. 2. Köhler curves for polyol particles. glycerol: black, erythritol: red, arabitol: orange, mannitol: yellow.
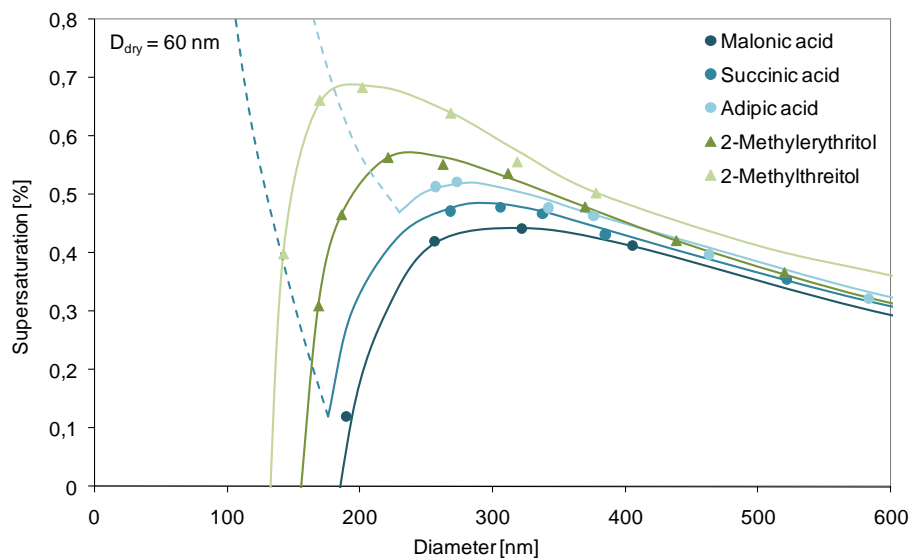
Fig. 3. Köhler curves for 2-methyltetrol and dicarboxylic acid particles. Triangles: Methyltetrols (2-methylthreitol: light green, 2-methylerythritol: dark green). Circles: dicarboxylic acids (malonic acid: dark blue, succinic acid: medium blue, adipic acid: light blue).
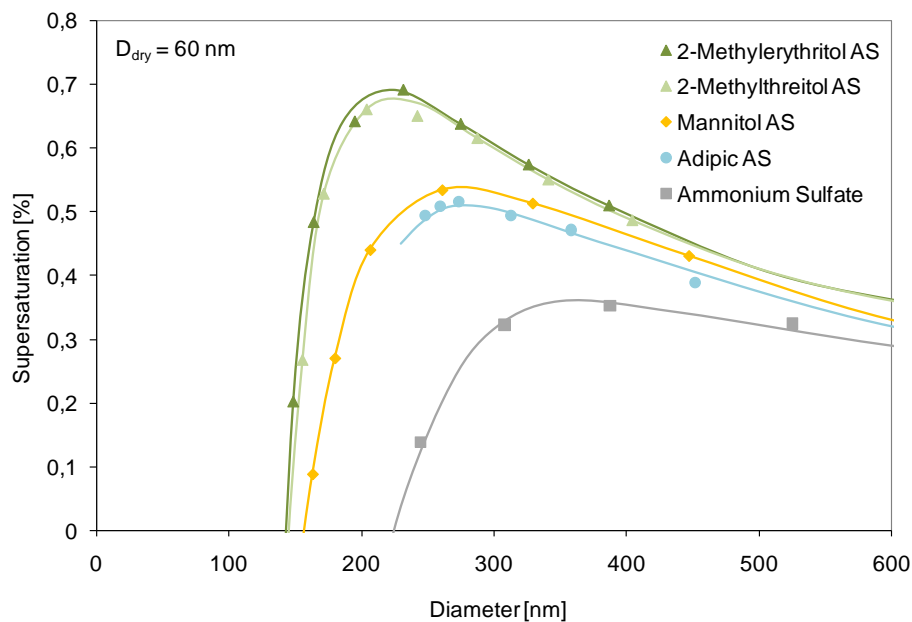
Fig. 4. Köhler curves for mixtures of organic compounds and sodium chloride. Grey squares: pure sodium chloride. Other compounds as in previous Figures.
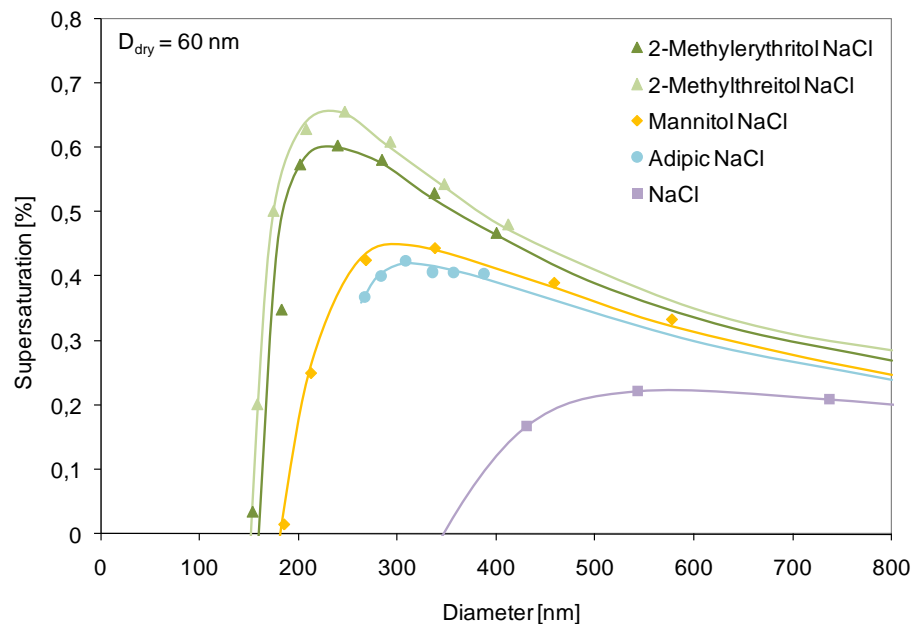
Fig. 5. Köhler curves for mixtures of organic compounds with ammonium sulphate. Purple squares: pure ammonium sulfate. Other compounds as in previous Figures.