The role of condensable vapours in atmospheric new particle growth and shrinkage

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New particle formation (NPF) is a key process that increases the number of aerosols in the atmosphere. It is characterized by a sudden burst of new particles of a few nanometres and subsequent growth to sizes of tens of nanometres. The latter growth is commonly considered as an irreversible process. However, particle shrinkage can occur due to the evaporation of the condensed species if the involved processes are reversible (Zhang *et al.*, 2012). To date, there are only a few observations of new particle shrinkage and thus still poorly understood.

In a multi-site NPF study in subtropical central Taiwan (Young *et al.*, 2012; Young *et al.*, 2013), 10-1000 nm particle number size distributions were measured with SMPS (GRIMM Model 5.500) in an urban (U), coastal (C), mountain (M) and downwind (D) area. The sampling campaigns were conducted during a cold and a warm season. Among the identified 14 NPF events, there were five particle shrinkage events during which the grown particles shrank back to sizes of ~10 nm, thereby creating a unique "arch-like" shape in the size distribution contour plot (Figure 1). The new particle growth and shrinkage rates (GR and SR), and their corresponding condensable vapour (C_v) and H_2SO_4 proxy (P) concentrations, and particle volume loss (VL) were estimated.

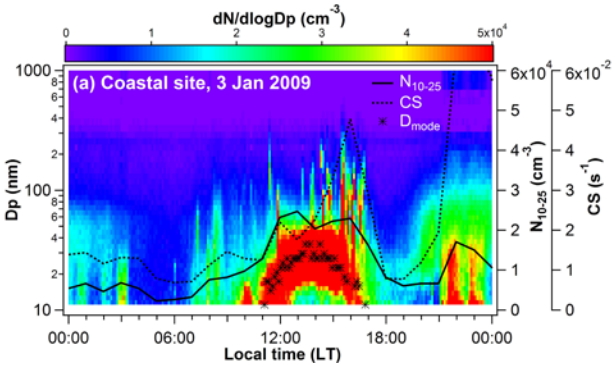

Figure 1. An example of new particle growth and shrinkage observed on the 3 January 2009.

Table 1 shows the GR, SR and GR-to-SR ratio, while Table 2 gives the C, P and VL for the five particle shrinkage events. As shown, the GR ranged from 7.4 to 17.3 nm hr^{-1}, whereas the SR ranged from 5.1 to 7.6 nm hr^{-1}. The SR-to-GR ratios were all above 0.44 indicating the SR were slower than the GR. The C of 1.69-6.59×10^8 cm^{-3} were required to explain the particle growth, whereas the P of 1.72-7.79×10^7 cm^{-3} were estimated to be produced under the observed atmospheric conditions. The P-to-C ratios were all below 0.22, suggesting that H_2SO_4 alone were not sufficient for the new particle growth. The VL on the 3 January 2009 and the 7 September 2010 were above 90%, indicating near complete evaporation. On the remaining three days, the VL were between 18% to 38%. These results clearly show that a notable fraction of the prior condensed chemical species evaporated from the particle phase to the gas phase. Bzdek *et al.* (2012) recently showed that sulfate contributed to 41-46% new particle growth, whereas NO_3^-, NH_4^+ and organics contributed to the remaining 54-59% growth. In particular, some of the latter chemical species are known to be semi-volatile and, as such, they have the potentials to evaporate off the new particles under favorable conditions.

Table 1. Growth rate (GR), shrinkage rate (SR) and SR-to-GR ratio during the new particle shrinkage events.

Date	Site	GR (nm hr^{-1})	SR (nm hr^{-1})	SR/GR
1/3/09	C	7.8	5.1	0.66
8/12/10	U	10.7	5.6	0.52
8/16/10	U	17.3	6.9	0.40
9/5/10	D	15.4	7.1	0.42
9/7/10	D	7.4	7.6	1.02

Table 2. Condensable vapour (C_v), H_2SO_4 proxy (P) concentrations, P-to-C ratio and particle volume loss (VL) during the new particle shrinkage events.

Date	Site	C (10^8 cm^{-3})	P (10^7 cm^{-3})	P/C	VL (%)
1/3/09	C	1.93	2.46	0.13	100
8/12/10	U	3.09	4.92	0.16	24
8/16/10	U	5.00	-	-	18
9/5/10	D	4.51	3.94	0.09	38
9/7/10	D	1.70	3.73	0.22	90

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