Reply for the referee no 3

We would like to thank the 3rd referee for reviewing the manuscript and making suggestions to improve the manuscript. We have made the following changes in the manuscript:

Referee: 1. It would be very informative to see at least an example of a nucleation event, which demonstrates a relatively small contribution of sub-3nm ions to the overall particle concentration during the event (p.1910, line 3 and following). Preferably, a quantitative value of ion contribution should be given.

Authors: We propose to make the following changes to the second paragraph of Sect. 5.2:

The nucleation events were observed on 86 % (DMPS), 84 % (negative ions) and 78 % (positive ions) of the analysed days at Marikana (Table 3, Fig. 9). With the ion spectrometer we were able to follow the growth of freshly nucleated ions, which allowed us to identify local charged particle formation events. The growing ion mode, however, was often visible from 3-7 nm upwards, which is an indication of a small contribution of ions to particle formation, or particle formation in residual layer or regional scale. If these observations were due to dominance of electrically neutral pathways in nucleation, then this would be in agreement with the theory and earlier observations that ion-mediated particle formation requires an environment of low nucleation rates, low temperature and aerosol content, but preferably having high concentrations of small ions and being rich with sulphuric acid (e.g. Laakso et al., 2002; Curtius et al., 2006; Iida 2006; Yu, 2010). Despite the relatively high estimate of sulphuric acid concentration, none of these requirements were met at Marikana. It is also possible that the nucleation already began in a residual layer before mixing into the boundary layer or some horizontal distance before the measurement site. Therefore, we suggest that regional new particle formation was significant at Marikana, despite the strong local pollution sources. This is understandable, as the typical transport time above the western BIC area is short (1-2 hours maximum).

Referee: 2. Likewise, the mentioned correlations between particle count in different size ranges with the measured gases, BC, CS, etc. (p.1907 last paragraph) should be supported either with a Table or graphs.

Authors: We agree with this comment and have implemented the following changes into the text and included a new table and figure in the manuscript:

Venter et al (2012) indicated that NO_x*, CO, BC and particulate mass mainly originated from local residential combustion for heating and cooking at Marikana, while* SO_2 *mainly originated from high stack industry emissions. Correlation coefficients between nucleation, Aitken and accumulation mode particle concentrations and trace gas, BC and CS are presented in Table 2. In addition, the relationship between particle and* SO_2 *concentrations are shown in Fig. 8. These observations together with the temporal variation of the particle number concentrations indicate that the morning and evening concentration peaks (Fig. 5) were due to the local residential combustion rather than due to industrial emissions. During the dry periods, which correspond to winter and the colder autumn and spring months, domestic burning is a major particle source. Residential combustion mainly takes place during the early morning and evening. Evening concentration peaks (Fig. 5) are usually more pronounced due to the need for space heating. The morning residential combustion period is usually curtailed due to warming temperatures and residents leaving for work. During the wet periods, which correspond to summer and the wetter autumn and spring months, domestic heating probably plays a smaller role as a particle source. Hence the observed Aitken and accumulation mode particles concentrations were higher in the dry periods (Fig. 4).*

Table 2. Here Pearson correlation coefficients (r) between various trace parameters and particle concentrations are presented. Particle concentrations were calculated for four size ranges: 12-20 nm, 20-50 nm, 50-100 nm and 100-840 nm.

Dry | Wet season	**Conc.**$_{12\text{-}20\ \mathrm{nm}}$	**Conc.**$_{20\text{-}50\ \mathrm{nm}}$	**Conc.**$_{50\text{-}100\ \mathrm{nm}}$	**Conc.**$_{100\text{-}840\ \mathrm{nm}}$
SO_2: daytime	0.48 | 0.49	0.38 | 0.51	0.18 | 0.36	0.29 | 0.30
night-time	-0.05 | -0.02	0.10 | 0.09	0.21 | 0.14	0.30 | 0.12
CO: daytime	-0.13 | -0.02	0.04 | -0.03	0.44 | 0.09	0.69 | 0.41
night-time	0.14 | 0.35	0.65 | 0.51	0.76 | 0.58	0.82 | 0.67
NO_x: daytime	-0.08 | 0.00	0.07 | -0.01	0.38 | 0.09	0.47 | 0.32
night-time	0.26 | 0.47	0.53 | 0.53	0.53 | 0.46	0.55 | 0.43
BC: daytime	-0.08 | 0.02	0.08 | -0.01	0.41 | 0.09	0.68 | 0.45
night-time	0.07 | 0.35	0.57 | 0.51	0.72 | 0.58	0.84 | 0.67
CS: daytime	0.02 | 0.15	0.30 | 0.40	0.74 | 0.72	0.96 | 0.86
night-time	0.00 | 0.12	0.68 | 0.46	0.91 | 0.85	0.99 | 0.97

Figure 8. Concentrations of nucleation ($dN_{12\text{-}20\ nm}$), Aitken ($dN_{20\text{-}50\ nm}$ and $dN_{50\text{-}100\ nm}$) and accumulation ($dN_{100\text{-}840\ nm}$) mode particles are presented as a function of SO_2 concentration during the dry season (upper panel) and the wet season (lower panel). Red dots indicate daytime concentrations (between 06:30-17:30) and black dots represent night-time (between 18:30-05:30) concentrations.