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Interactive comment on "Characterization of aerosol particle episodes in Finland caused by wildfires in Eastern Europe" *by* J. V. Niemi et al.

J. V. Niemi et al.

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We thank the referees for their constructive reviews and suggestions. We will respond here to the comments from the both referees individually.

RESPONSE TO THE COMMENTS OF REFEREE #1:

Individual particle analysis (IPA):

We revised the SEM/EDX sections of the paper based on the referee's comments. We present more clearly that due to agglomeration of particles, only major changes in aerosols during the episodes are observed (especially elevated K/S ratio, and relative increase of coarse Ca-rich particles). We also present more clearly that the elemental results are semi-quantitative and they are used to describe changes in the relative

ratios of elements. They are not absolute weight percentages. We added S/K ratio values to show the relative changes more clearly. More detailed response on the referee's comments on IPA is shown below.

The substrate for IPA sampling was fibreglass filter tape which is (already) mentioned in section 2.4 (p. 2474, line 13). We agree that the sampling method and substrate used is not optimal for individual particle SEM/EDX analysis. The ideal particle samples for SEM/EDX analysis are collected on smooth substrate and particles do not overlap with each others (no overload). However, that kind of IPA samples are usually collected only during specific field campaigns, and thus ideal samples are seldom available during unusual events. The particle samples collected during ordinary long-term air quality monitoring measurements are available also during episodic events. Those samples offer possibility to analyze chemical composition of particles although they are not ideal for analysis.

One of the common particle mass monitor types is Eberline FH 62 I-R sampler (Eberline Instruments) which collects particles typically on fibreglass filter tape. Particles are collected on the small rounded area (diameter ˜1.5 cm) and the sampling time is typically one day. SEM/EDX method is obviously one of the best methods to analyse these samples because only small amount of particles if sufficient for analysis, and the method provides information on the elemental composition and morphology.

Before SEM/EDX analysis, particles are transferred from the fibreglass filter to adhesive tape (mounted on the aluminium stub) by pressing lightly the surface of fibreglass filter against the surface of adhesive tape. Particles and also few fibres from fibreglass filter are attached on the adhesive tape. It is possible that this transfer method may cause bias on particle populations. This is the main reason why we use the random selection of particles during SEM/EDX analysis. It guarantees, that particles are analysed from a large area and not only from a selected small area which might be biased. The location of analysis area is randomly changed after every analysis. During the random selection of particles the magnification of SEM is always set as 2000. Each

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time, the particle or agglomerate located most close to the small point mark (visible in the SEM monitor) is selected for analysis. This procedure guarantees as random particle selection as possible for the fibreglass filter samples. If the method suggested by the referee is used ("to analyze all particles in the selected field") the bias would probably be greater than with our method: one field might contain the whole population (100) of analyzed particles meaning that only a very small area of the sample would be included. Furthermore, all analyses are performed by the same person in each study to minimize possible subjectivity in analysis. In the present study, J.V. Niemi analyzed all particles. He also analyzed all particles for the March 2002 episode study with the same devices and methods, which guarantees especially reliable comparison of these two studies.

The agglomeration of particles during sampling and also during sample preparation for SEM/EDX analysis limits possibility to analyse individual fine particles. Therefore, we analyzed both individual particles and agglomerates (formed mainly from submicron particles). The focus of the analysis is to find major changes in aerosols during different episodes. We have analyzed with SEM/EDX-method hundreds of particle samples during the last years. These studies have focused on the composition and origin of particles during different peak particle concentration episodes (e.g. Tervahattu et al., 2002; Niemi et al., 2004; Tervahattu et al., 2004). During these studies we have got confidence that our sampling and analysis method suits well for the characterization of major changes in aerosol composition (but it does not suit well if the focus is to analyse detailed changes in fine aerosol populations). The SEM/EDX elemental results are semi-quantitative, and they do not describe absolute weight percentages (ZAF-corrected relative weight percentages, analyzed elements normalized to 100%). The results are used to compare relative changes in the ratios of elements analyzed (not absolute values!). This is already mentioned in the paper but in the revised text it is presented more clearly.

Specific comments:

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1. Title: We already removed “PM2.5 episodes” from the title based on the editor’s comments and replaced it with “aerosol particle episodes”. The opinions of referee #1, referee #2 and editor are slightly different. Thus we consider that the current topic (including the expression “aerosol particle episodes”) might be suitable.

2. Abstract: We changed the expression for the SEM/EDX method to “scanning electron microscopy (SEM) coupled with energy dispersive X-ray microanalyses (EDX)”.

3. We checked the use of grammar and spelling.

RESPONSE TO THE COMMENTS OF REFEREE #2:

Referee #2 writes in the review that ”The authors identify the ratio of Accumulation mode number to Aitken mode number as a key microphysical indicator for biomass burning aerosol”. We did not mention that in our paper but we suggested that high values of accumulation mode to Aitken mode number concentration ratios might be a good indicator of LRT episodes in generally. We changed the text of article so that this is more clearly expressed.

We checked the use of grammar and spelling.

Specific comments:

1. Title: The same comments as above for referee #1

2. Abstract: The same comments as above for referee #1.

3. Section 2.2: We added more detailed description of the sampling protocols including sampling heights (2 m). Furthermore, the details of the measurement methods are also available in the references mentioned in the article: (Laakso et al., 2003, mass fraction measurements with the impactor) and (Aalto et al., 2001, number size distributions with the DMPS).

4. Section 3.2: We prepared typical (mean) number size distributions for the episodes and for reference period as suggested by the referee. These number size distribu-

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tions show clearly that during the episodes the concentrations of accumulation mode (90-500 nm) particles increased while the concentrations of nucleation (3-25 nm) and Aitken mode (25-90 nm) particles decreased. In our opinion, these changes are already shown clearly enough in Table 2 as well as in Figs. 6a and 6b, and we feel that it is not be necessary to involve this new figure to the article. However, this figure will be shown in the response letter to the editor.

We did not mention in the text that "Aitken mode particles are not subject to long-range transport" but we wrote that "The Aitken mode particles cannot be transported in the atmosphere as far away as accumulation mode particles". However, the long-range transport of Aitken mode particle is limited due to their short life time (especially when particle concentrations are high, e.g. due to strong long LRT episode). E.g. if we assume, that background distribution has 1000 particles/cc, having diameter of 100 nm and our Aitken mode particles have diameter of 50 nm, coagulation coefficient between these particles is about 1e-7. Thus characteristic lifetime of smaller particles is 1/(1e-7*1000) which is about 1e4 seconds ˜ 3 hours. As a result of coagulation, we loose Aitken mode particle, but we still have an accumulation mode particle, now with diameter of 104 nm. In case of accumulation mode particles, this characteristic lifetime is longer.

Additionally, Aitken mode particle grow to larger sized due to condensation and are also more efficiently scavenged due to wet and dry deposition compared to accumulation mode particles. The characteristic life time for accumulation mode particles is much longer because scavenging processes are less efficient and condensation is not able to change particle sized above accumulation mode size fast enough. Suitable references for above mentioned statements are e.g. Seinfeld and Pandis (1998, p. 100, 662, 971, 1020) and Mönkkönen et al. (2004). We did not find suitable reference to show the transport distances of Aitken particles in different atmospheric conditions. However, in our opinion, the short lifetime of Aitken particles under polluted conditions is sufficient evidence to support the conclusions we made.

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5. Section 3.3: We added discussion on the statistical significance of the effects of LRT on the ratio of the ion sum to PM10 mass (related to Fig. 8). The differences between the episodes and the reference period were statistically significant ($p<0.001$, One Sample T-test, SPSS 10.0 statistical program).

6. We added more detailed axis titles for Fig. 2 and 7, and we added axes title for Fig. 6b.

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