Supplementary information for:

Characterisation of the filter inlet system on the BAE-146 research aircraft and its use for size resolved aerosol composition measurements

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SEM aerosol particle classification scheme

The classification was done within the AZtecFeature software by Oxford instruments. The software allows the user to create a custom made categorisation scheme based on the chemical and morphological properties of the features detected by the software. Each particle is tested against a set of rules in order to categorise them (we introduce the flow chart of rules below). In this study the 32 raw categories are simplified into 10 atmospherically relevant categories. For a particular dataset, the number of categories could be increased or decreased if necessary according to the characteristics of the sample.

The software calculates the weight percentage (wt %) of each detected element with its statistical error (σ). In our classification scheme, we have imposed that all the detected elements must be statistically significant in order to be considered as present. In order to be statistically significant, the wt % needs to be a certain confidence level above the σ. We explored the appropriate value of sigma for our application below.

Our analysis is distinguished from others in the literature in that we use a relatively thick Ir coating (30nm) as well as a relatively low EDS integration time in order to get data from many particles in a session. Some of the secondary EDS peaks of Ir overlap in some cases with some of the atmospherically relevant elements (the primary peak does not). This produces some issues like a larger σ in some elements. This effect is quite noticeable for Al and S, where some clear peaks of these elements were not statistically significant at a confidence level of 3. In the figure SI 1 we show the results of a test where we studied the effect of changing the confidence level from 3 to 2 σ in the particle categorisation carried out by the classification scheme. The only effect of this change yields on the Al and S. When going from 3 to 2 σ as a confidence level, more Al is detected in the sample, so some Si-rich particles (from rule 25) are detected as Al-Si rich particles (rule 5) instead. Manual inspection of a subset of these particles revealed that the Al peak that wasn't being identified at 3 σ is an actual Al signal that was detected at 2 σ. Likewise, some significant S peaks were not being detected at a confidence level of 3 σ but they were at 2 σ, leading to more S rich particles (rule 14) that were labelled as Other from the rule 32 at a higher confidence level. The variation in the confidence level didn't modify the number of particles in other categories, so we recommend to use a 2 σ value in order to minimise the underestimation of Al-Si and S rich particles.

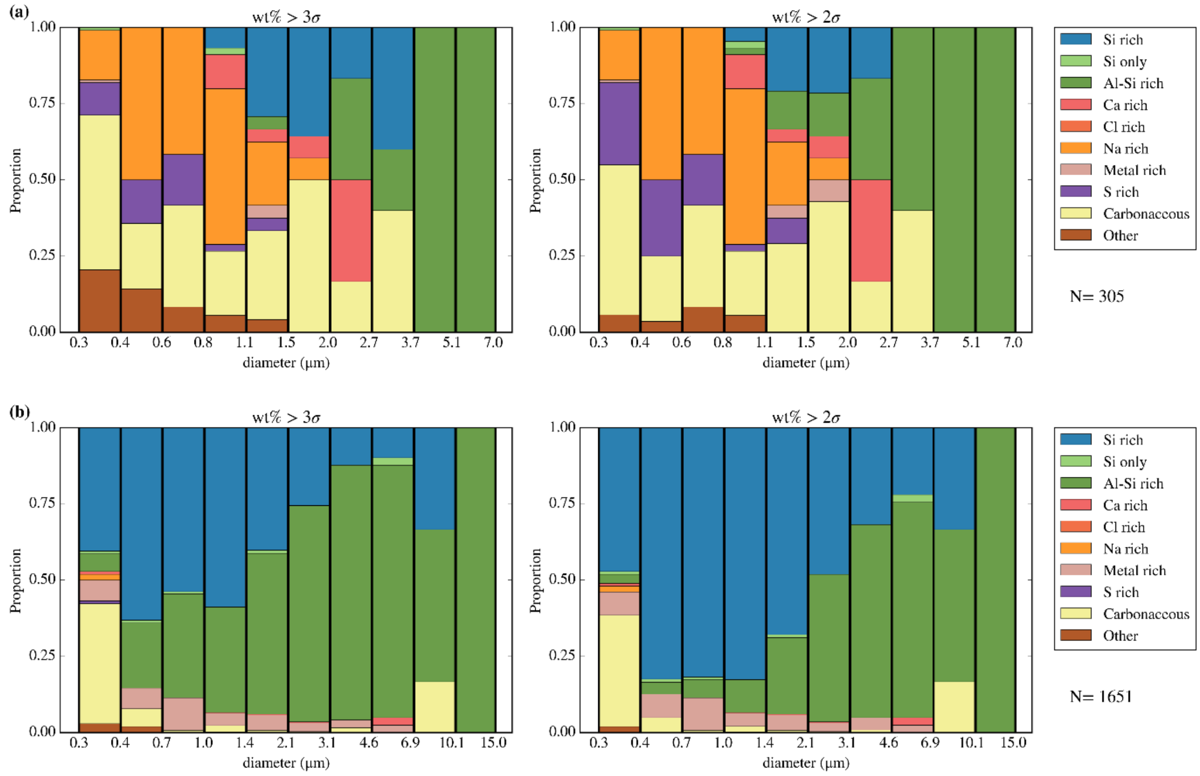


Figure SI 1. Size-segregated composition of two aerosol samples for different element detection confidence levels. The samples are 2018/03/18 from 19:28 to 19:48 UTC in north Alaska (a) and 2017/10/02 from 16:24 to 16:40 UTC in Iceland (b). The two samples are very different since the first sample presented a very low aerosol loading and it is dominated by Na rich particles, Carbonaceous and mineral origin aerosol (Si rich, Si only, Al-Si rich) with significant contributions of S rich particles whereas the second sample presented a high aerosol loading and it was mainly dominated by mineral origin aerosol. The different in the confidence mainly affected the Si and Al-Si rich particles as well as the S rich particles in the sample (a), whereas it only affected the Si and Al-Si rich particles in the sample (b).

For some unclear reason, the secondary peaks of the Ir peak can be mislabelled as minor (but detectable) concentrations of other elements as Si and Cu. We noticed that these EDS peaks coming from random places of the filter, not only aerosol particles. As a consequence, we observed that a carbonaceous particle (especially the small ones) could be wrongly labelled as Si only or Metal rich (Cu). Therefore, we added some rules in the classification scheme to avoid this problems (rules 2 and 28). These rules may not work for all the Si and Cu artefacts, and they may also hide some actual signals of Si and Cu coming from aerosol particles, but adding them creates a more representative analysis.

As mentioned in the Sect. 4, all the Cr dominated particles were removed from the analysis since they are very likely to be artefacts from the filter as one can see in the Fig. SI2, where the size-resolved composition of blank and handling blank filters is presented. For an individual case in which Cr is a very frequent element, they could be included if necessary.

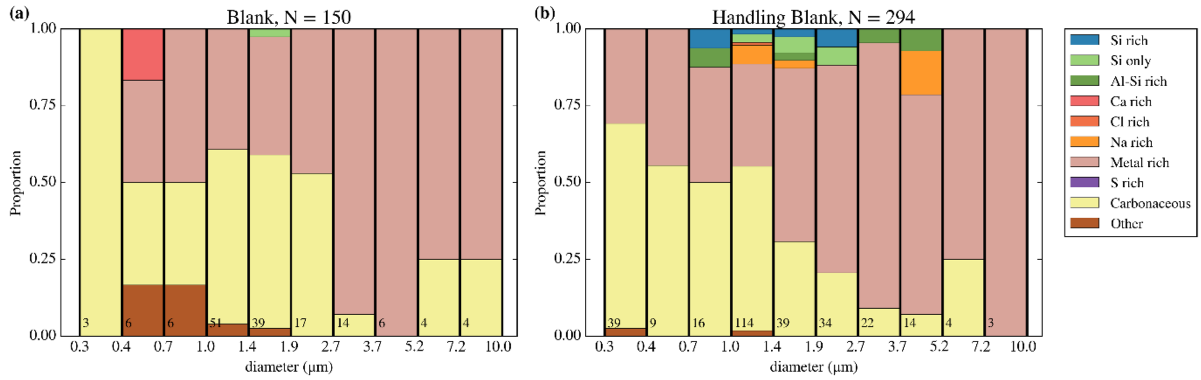
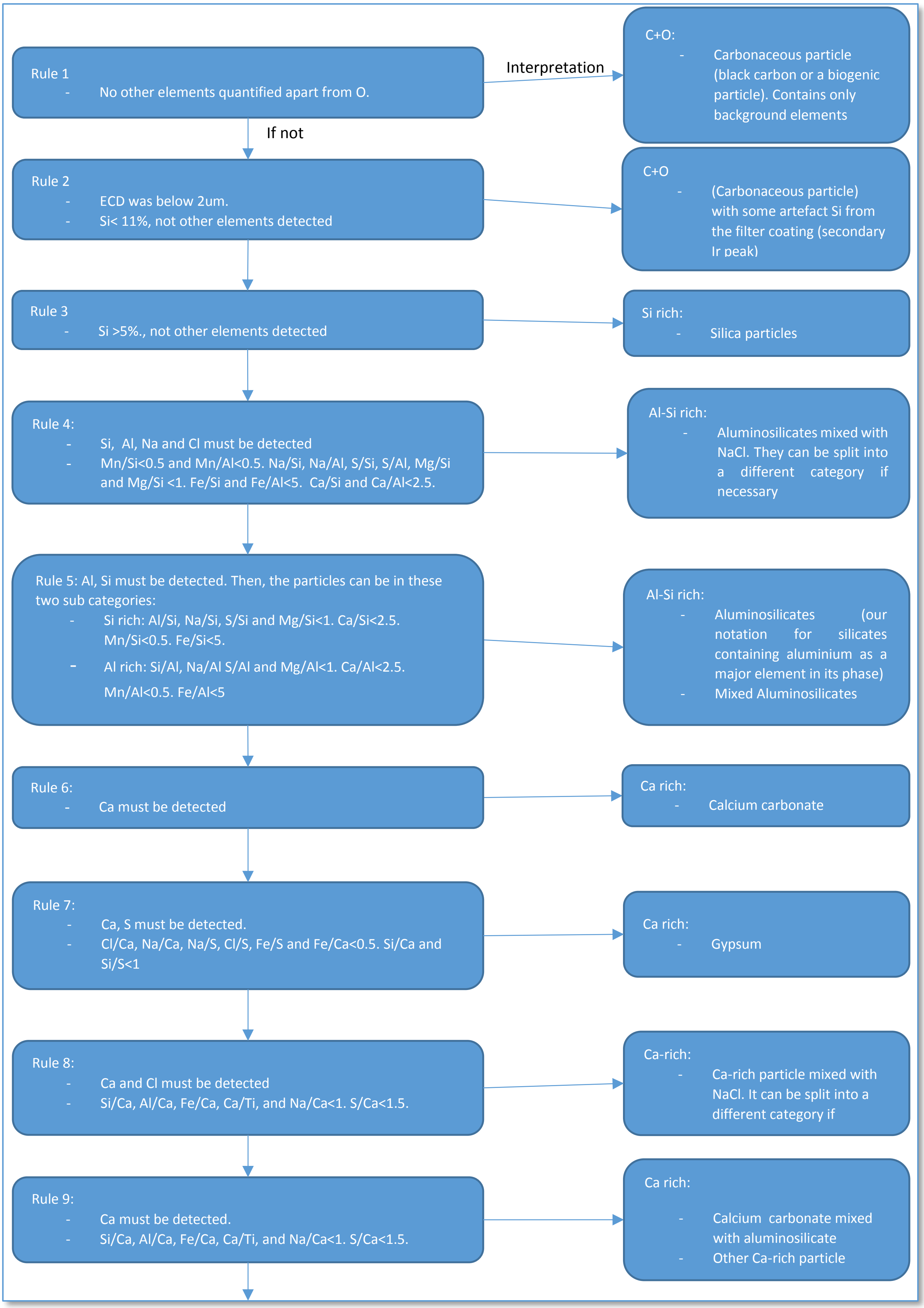
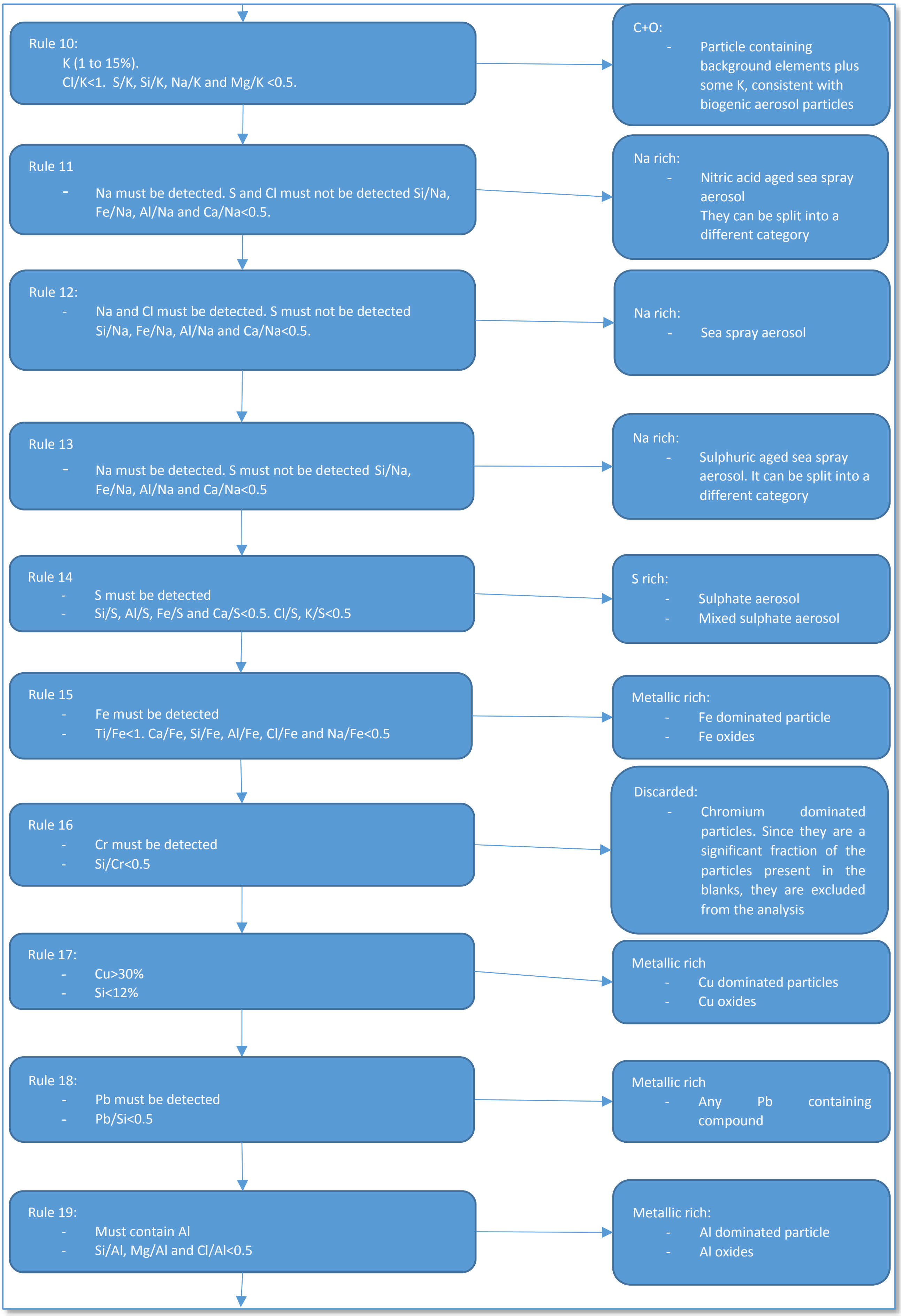


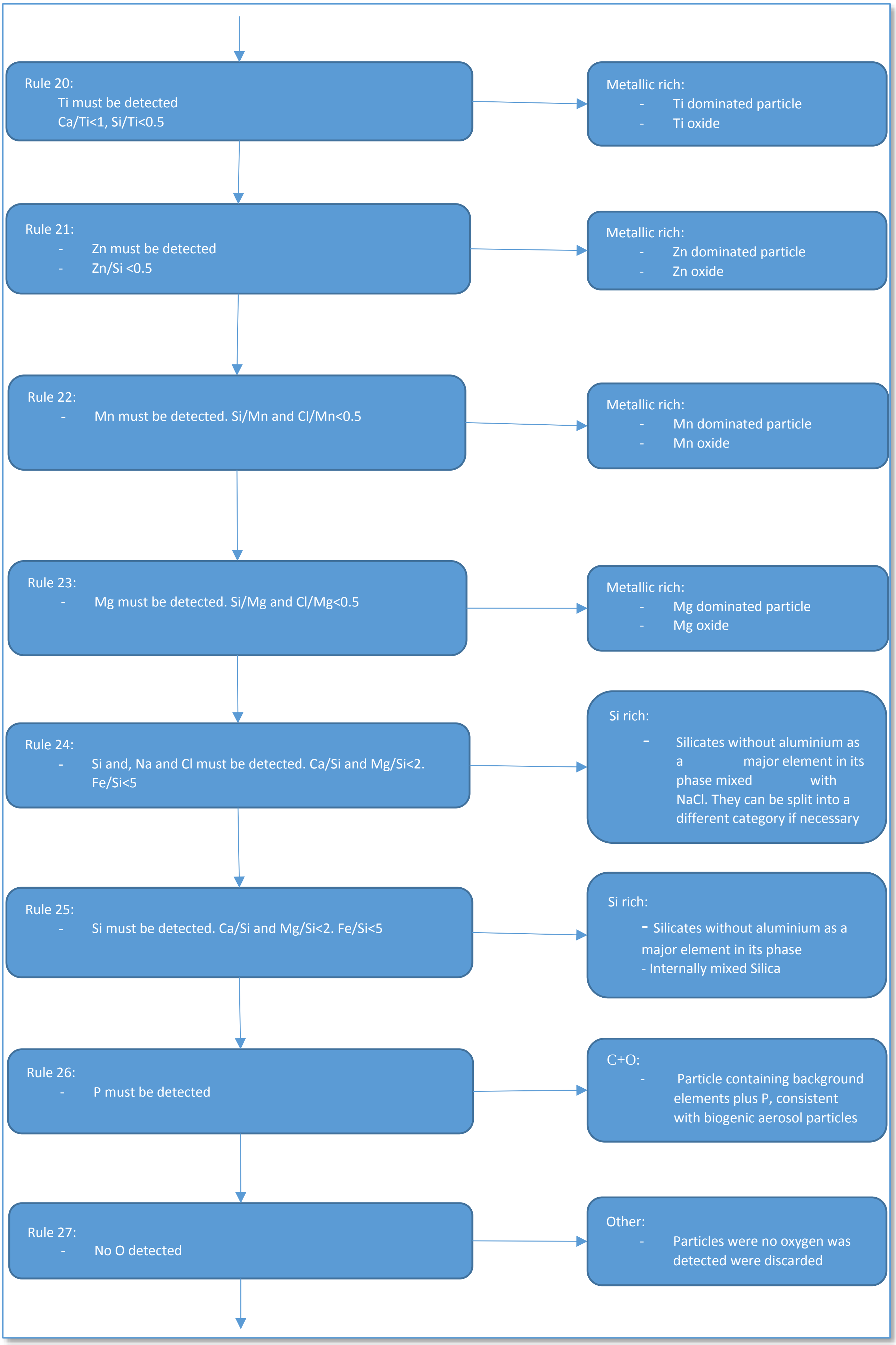
Fig. S2. Size segregated composition of the artefact particles found in 3 blank filters (a) and a handling blank filter (b). The number of particles analysed in each case appears in each image. Almost all the particles present in the Metallic rich category (97 and 96 % respectively) were Cr rich particles.

In Fig. S3 one can see the classification scheme. C and Ir are present in the filter material and coating at all locations on the filter, therefore these elements have been excluded from the classification analysis. O is also a background element, but has been included to aid particle identification. Elemental totals (excluding C and Ir) were normalised to 100% and then classified by AZtecFeature using the rules described in the figure SI 3. Even though it is not stated in each rule, O detection was a requirement in all the rules. If the morphological and chemical properties of a particle match with a particular rule in the scheme, the particle is labelled with that rule. One or more rules can be summarised as a category when plotting the data. We have summarised all the rules into 10 categories, which were explained in Sect. 7 of the main text. An interpretation of the type of aerosol particle for each rule is also given in the table, as well as the final category it belongs to. The number of categories can be changed if the conditions of the sample need it, for example, Al-Si rich particles could be split into Al-Si rich particles containing Na+Cl (rule 4) and Al-Si rich particles not containing Na+Cl (rule 5). This would be interesting, for example if studying the mixing of mineral dust with sea spray aerosol in a particular environment.


Rule 1
- No other elements quantified apart from O.
Interpretation
C+O:
- Carbonaceous particle (black carbon or a biogenic particle). Contains only background elements
If not
Rule 2
- ECD was below 2um.
- Si< 11%, not other elements detected
C+O
- (Carbonaceous particle) with some artefact Si from the filter coating (secondary Ir peak)
Rule 3
- Si >5%., not other elements detected
Si rich:
- Silica particles
Rule 4:
- Si, Al, Na and Cl must be detected
- Mn/Si<0.5 and Mn/Al<0.5. Na/Si, Na/Al, S/Si, S/Al, Mg/Si and Mg/Si <1. Fe/Si and Fe/Al<5. Ca/Si and Ca/Al<2.5.
Al-Si rich:
- Aluminosilicates mixed with NaCl. They can be split into a different category if necessary
Rule 5: Al, Si must be detected. Then, the particles can be in these two sub categories:
- Si rich: Al/Si, Na/Si, S/Si and Mg/Si<1. Ca/Si<2.5. Mn/Si<0.5. Fe/Si<5.
- Al rich: Si/Al, Na/Al S/Al and Mg/Al<1. Ca/Al<2.5. Mn/Al<0.5. Fe/Al<5
Al-Si rich:
- Aluminosilicates (our notation for silicates containing aluminium as a major element in its phase)
- Mixed Aluminosilicates
Rule 6:
- Ca must be detected
Ca rich:
- Calcium carbonate
Rule 7:
- Ca, S must be detected.
- Cl/Ca, Na/Ca, Na/S, Cl/S, Fe/S and Fe/Ca<0.5. Si/Ca and Si/S<1
Ca rich:
- Gypsum
Rule 8:
- Ca and Cl must be detected
- Si/Ca, Al/Ca, Fe/Ca, Ca/Ti, and Na/Ca<1. S/Ca<1.5.
Ca-rich:
- Ca-rich particle mixed with NaCl. It can be split into a different category if
Rule 9:
- Ca must be detected.
- Si/Ca, Al/Ca, Fe/Ca, Ca/Ti, and Na/Ca<1. S/Ca<1.5.
Ca rich:
- Calcium carbonate mixed with aluminosilicate
- Other Ca-rich particle



Rule 10:
K (1 to 15%).
Cl/K<1. S/K, Si/K, Na/K and Mg/K <0.5.
C+O:
- Particle containing background elements plus some K, consistent with biogenic aerosol particles
Rule 11
- Na must be detected. S and Cl must not be detected Si/Na, Fe/Na, Al/Na and Ca/Na<0.5.
Na rich:
- Nitric acid aged sea spray aerosol
They can be split into a different category
Rule 12:
- Na and Cl must be detected. S must not be detected Si/Na, Fe/Na, Al/Na and Ca/Na<0.5.
Na rich:
- Sea spray aerosol
Rule 13
- Na must be detected. S must not be detected Si/Na, Fe/Na, Al/Na and Ca/Na<0.5
Na rich:
- Sulphuric aged sea spray aerosol. It can be split into a different category
Rule 14
- S must be detected
- Si/S, Al/S, Fe/S and Ca/S<0.5. Cl/S, K/S<0.5
S rich:
- Sulphate aerosol
- Mixed sulphate aerosol
Rule 15
- Fe must be detected
- Ti/Fe<1. Ca/Fe, Si/Fe, Al/Fe, Cl/Fe and Na/Fe<0.5
Metallic rich:
- Fe dominated particle
- Fe oxides
Rule 16
- Cr must be detected
- Si/Cr<0.5
Discarded:
- Chromium dominated particles. Since they are a significant fraction of the particles present in the blanks, they are excluded from the analysis
Rule 17:
- Cu>30%
- Si<12%
Metallic rich
- Cu dominated particles
- Cu oxides
Rule 18:
- Pb must be detected
- Pb/Si<0.5
Metallic rich
- Any Pb containing compound
Rule 19:
- Must contain Al
- Si/Al, Mg/Al and Cl/Al<0.5
Metallic rich:
- Al dominated particle
- Al oxides


Rule 20:
Ti must be detected
Ca/Ti<1, Si/Ti<0.5
Metallic rich:
- Ti dominated particle
- Ti oxide
Rule 21:
- Zn must be detected
- Zn/Si <0.5
Metallic rich:
- Zn dominated particle
- Zn oxide
Rule 22:
- Mn must be detected. Si/Mn and Cl/Mn<0.5
Metallic rich:
- Mn dominated particle
- Mn oxide
Rule 23:
- Mg must be detected. Si/Mg and Cl/Mg<0.5
Metallic rich:
- Mg dominated particle
- Mg oxide
Rule 24:
- Si and, Na and Cl must be detected. Ca/Si and Mg/Si<2. Fe/Si<5
Si rich:
- Silicates without aluminium as a major element in its phase mixed with NaCl. They can be split into a different category if necessary
Rule 25:
- Si must be detected. Ca/Si and Mg/Si<2. Fe/Si<5
Si rich:
- Silicates without aluminium as a major element in its phase
- Internally mixed Silica
Rule 26:
- P must be detected
C+O:
- Particle containing background elements plus P, consistent with biogenic aerosol particles
Rule 27:
- No O detected
Other:
- Particles were no oxygen was detected were discarded

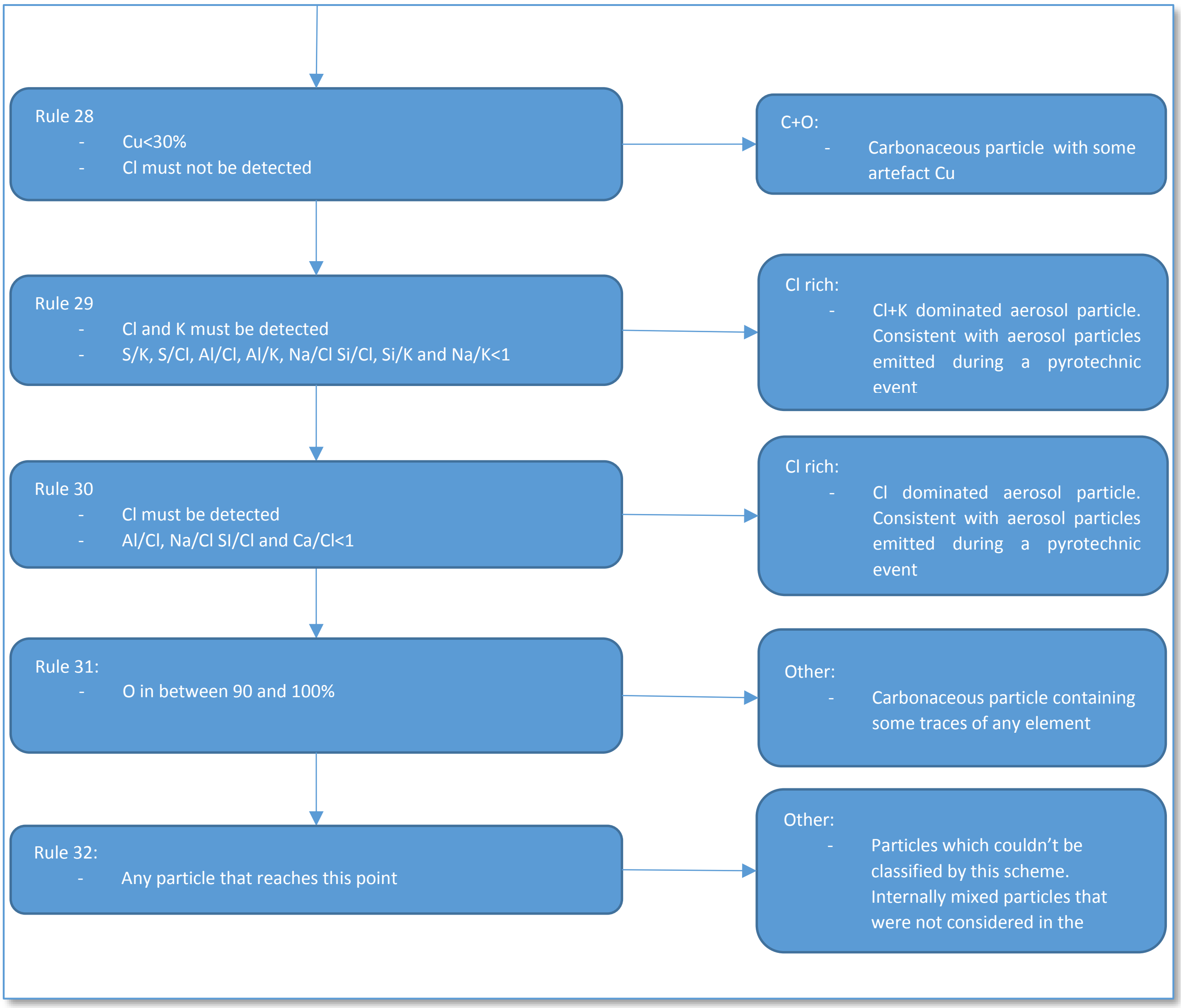


Figure S3. Description of the classification scheme. The 32 sets of rules used to categorise aerosol particles can be seen in a descendant order. C and Ir was excluded from all the particles for this analysis. In spite of not being mentioned, presence of O was required in all the sets of rules.