Interactive comment on “Anthropogenic and natural constituents in particulate matter in the Netherlands” by E. P. Weijers et al.

Anonymous Referee #1
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This manuscript presents a detailed analysis of the chemical composition of PM10, PM2.5 and PM10-2.5 at 5 sites in the Netherlands. It is well written and the results are interesting, even though conclusions regarding the natural fraction in PM10 and PM2.5 seem somewhat excessive. This is my main concern. The methodology applied is not especially novel, but the work is very comprehensive and exhaustive in the details.

Page 26516, line 4: it would be advisable to use a different acronym for CBM, to avoid confusion with the Chemical Mass Balance receptor model. Please insert “pragmatic” after “the so-called”. Page 16517, line 12: what is DCMR? Page 26517, line 19: were mineral matter components (Ca, Al, Mg, K, Fe, ...) analysed? Page 26518, line 3: were both types of filters intercompared in terms of mass? Page 26518, line 5: how was the soluble fraction extracted from the filters? Please describe the methodology used.

Page 26519, line 8: please list the elements determined by ICP-MS. Same for the elements summed up to form “total metals”. Figure 3 shows a rather large NA mass which is probably mineral matter given that the major tracers (Al, Ca, ...) were not determined. Is this correct? Otherwise please describe how these constituents were analysed. Page 26523, line 5: it would be useful for the authors to briefly discuss the limitations of applying a constant factor of 1.3 to TC, instead of converting OC to OM, based on their results. They obtain the “maximum change” at the kerbside site, despite the fact that at this site TC is made up of a larger proportion of EC than OC due to the fresh vehicular emissions, and therefore the increase in the mass due to heteroatoms should be lower than at a rural site where the proportion of OC is higher. Table 4: please provide values also as % of mass, this would greatly aid the reader. Page 26523, line 25: The levels of SS in PM10 in Cabauw are surprisingly low, especially in comparison with those at Hellendoorn and Vredepeel (located farther away from the coastline). How do the authors interpret this? Page 26525, line 5: the absence of an increase in mineral matter seems unusual, given that the authors do detect an increase in the metal content. The metals are deposited on the traffic ways and therefore should be resuspended simultaneously with mineral dust from the pavement. If no increase in the metal content had been detected, it could have been argued that due to the frequent rain and humidity road dust resuspension is not a significant source in Rotterdam. However, given that metals were detected, it seems unlikely that they would be dissociated from mineral matter originating from the pavement. How do the authors explain this? Would they expect to find the same results in other major cities, e.g., Amsterdam? Page 26526, section 4.2: this section should be summarised, the role of water uptake by aerosols in the mass closure is already well known. Page 26530, line 17: based on the authors’ reasoning, the contribution of natural sulphate should be 5%, not 5-10%. In addition, considering the vast contribution of total nitrate to PM levels in the Netherlands, 5% seems like a rather large % for natural nitrate contributions. The authors’ reasonings in this entire section are perfectly valid from a theoretical point of view, but it would be interesting to read reviewers’ comments if a study originating in Southern Europe ever tried to justify sulphate and nitrate as being natural... Regarding
OC, as stated by the authors the debate on the natural vs anthropogenic sources of OM is currently large and unconclusive. I would therefore recommend a more conservative approach and to remove OC from the calculation of the natural PM fraction. Page 26532, line 10: it was stated in previous sections that road dust resuspension in Rotterdam is not linked with mineral dust, which is a contradiction with what is stated here. Page 26532, line 26: 20% of mineral dust as natural in NL is not a very conservative approach. Page 26533, line 15: in general, the assumptions described in section 4.4 regarding the natural fraction of PM seem to provide a rather high estimation (25% of PM10). The approach is correct, but more conservative assumptions would probably yield a more realistic estimate.

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