Comments from Referee #1

This manuscript continues to be rather poor in English writing. Thus, making it difficult to understand and provide constructive comments. The "Abstract" is very confusing and difficult to follow with statements like "(denuded vs. undenuded)" and "frequencies (24 vs 48 h averaged)". It is not clear what is "operationally defined value of the thermal optical method"? There are number of such confusing statements. A disagreement of 15% is well within the analytical uncertainty of EC measurement on thermal-optical analyzer. It sounds rather absurd to refer 24 h sampling as low frequency sampling. Overall the manuscript needs substantial revision and improvement in English writing. It lacks the desired level of scientific merit and unacceptable in its present form.

Our Response:

The manuscript has been revised carefully and then polished by Dr. Guenter Engling at the Desert Research Institute. In the revised version, the abstract and conclusions have been completely re-written; and moreover, substantial changes have been made to the introduction (now the background information is introduced more clearly). We think the revised manuscript should be much easier to follow.

Moreover, a diagram and two tables are presented at the end of this response, which are expected to be useful for the referee to review the revised manuscript. The diagram (Figure R1) describes the design of the present study; key observational results and corresponding conclusions are summarized in Table R1; statistical results associated with the major conclusions of this study are presented in Table R2.

In addition, the referee mentioned several specific points regarding the abstract, including (1) the abstract is very confusing and difficult to follow with statements like "(denuded vs. undenuded)" and "frequencies (24 vs 48 h averaged)", and (2) it is not clear what is operationally defined value of the thermal-optical method. These types of statements are avoided in the revised abstract.

The referee also mentioned that a disagreement of 15% is well within the analytical uncertainty of EC measurement on thermal-optical analyzer (this comments is associated with the result that EC concentrations of 48 h averaged samples were about 15% lower than results from 24 h averaged ones). It was estimated that the precision of EC analysis was within 5% for this study. The precision was evaluated as the ratio of the standard deviation of the duplicate measurements to the average value. A precision

of 5% is comparable with results from previous studies based on the DRI carbon analyzer (e.g., Chow et al., 2004). On the other hand, statistical analysis suggested that the difference in EC results between 48 and 24 h averaged samples was significant at a 95% level of confidence (2 tailed p = 0.000; Table R2).

Finally, the referee pointed out that it sounds rather absurd to refer 24 h sampling as high frequency sampling. In this study, samples from channel 1 and 2, which are collected at a relatively high frequency, are referred to as high frequency samples for simplicity; and correspondingly, samples from channel 3 are termed low frequency samples. This does not necessarily mean that 24 h averaged sampling should be considered as high frequency sampling elsewhere. In the revised manuscript, this pointed is clarified in Table 1.

Reference

Chow, J. C., Watson, J. G., Chen, L. W. A., Arnott, W. P., and Moosmüller, H.: Equivalence of elemental carbon by thermal/optical reflectance and transmittance with different temperature protocols, Environ. Sci. Technol., 38, 4414–4422, 2004.



Measured parameters include total carbon (TC), organic carbon (OC), elemental carbon (EC), and optical attenuation (ATN).

This study investigates the influence of sampling frequency on the measurement of carbonaceous aerosol, with a focus on the uncertainties associated with sampling and thermal-optical analysis including:

- the positive sampling artifact caused by the adsorption of gaseous organics by the commonly-used quartz filter (which tends to overestimate OC concentrations)
- the negative sampling artifact due to the evaporation of the collected particles (which tends to underestimate OC concentrations)
- the analytical artifact of thermal-optical methods caused by the transformation of OC into char OC (which tends to underestimate EC and thus overestimate OC)
- the shadowing effect in the determination of ATN which means an increased underestimation of ATN with increasing filter loadings

Comparisons are made based on 48 h averaged concentrations for TC, OC and EC, whereas comparisons are based on 48 h integrated values for ATN.

Results from the denuded quartz filter in channel 1 (DQ) are used as the reference values.

For simplicity, samples from channel 1 and 2, which are collected at a relatively high frequency, are referred to as high frequency samples; and correspondingly, samples from channel 3 are termed low frequency samples. This does not necessarily mean that 24 h averaged sampling should be considered as high frequency sampling elsewhere.

Figure R1. Diagram of the design of this study.

Table R1. Key observational results and corresponding conclusions:

Observational results	Corresponding conclusions	
TC measured by the low frequency BQ (in channel 3) could be lower than that measured by the high frequency DQ (in channel 1). This phenomenon is not apparently associated with filter loading, instead, is observed only during a distinct period characterized by high humidity. (Section 3.1)	(1) The negative sampling artifact of a bare quartz filter could be remarkably enhanced due to the uptake of water vapor by the filter medium.	
EC concentrations of the low frequency BQ (in channel 3) are about 15% lower than results from the high frequency samples (i.e., DQ in channel 1). (Section 3.2)	(2) The analytical artifact is more significant for the low frequency samples.	
48 h integrated ATN is about 10% lower for the low frequency BQ (in channel 3) compared to the high frequency samples (i.e., DQ in channel 1). (Section 3.3)	(3) The shadowing effect in the determination of ATN is more considerable for the low frequency samples.	
The EC _R (EC defined by the reflectance charring correction) to EC _T (EC defined by the transmittance charring correction) ratios are much higher for the low frequency BQ (in channel 3) compared to the high frequency samples (i.e., DQ in channel 1). (Section 3.4)	(4) EC results of the low frequency samples depend more strongly on the charring correction method.	

Table R2. Statistical results for the key comparisons included in this study (2-tailed p < 0.1 indicates significant difference at a 95% level of confidence, whereas 2-tailed p > 0.1 indicates insignificant difference). Results shown in Table R2 are also presented in the Supplement.

Y	Х	2-tailed <i>p</i>	Corresponding conclusions
Low-frequency TC_{BQ} during the high RH period	High-frequency TC_{DQ} during the high RH period	0.044 (Paired t-test)	(1)
Low-frequency EC _{BQ}	High-frequency EC _{DQ}	0.000 (Paired t-test)	(2)
Low-frequency ATN _{BQ}	Integrated high-frequency ATN _{DQ}	0.000 (Paired t-test)	(3)
EC_R to EC_T ratios of the low frequency samples	EC_R to EC_T ratios of the high frequency, denuded samples	0.005 (Independent t-test)	(4)