

Reply to Editor

We thank the Editor for the positive feedback and bringing forth the points that really needed clarification. Our answers below are written by blue color. The changes made in the manuscript are written below by red color.

Thank you for the thoughtful responses and edits. After reviewing the responses, there are only 2 small questions before acceptance.

Editor question: Do you feel the following point is adequately conveyed in the manuscript to avoid similar confusion from future readers of the paper (and referenced to help any readers find the details)?

“page 13, line 395: “we calculated the expected statistical variability using the Co-spectrum”. What does this mean - haven’t heard of this technique before?

The following clarification was added on page 13-14, lines 399-403:

The statistical variability of the covariance (which equals the integral of the Co-spectrum) depends on the shape of the Co-spectrum. A white Co-spectrum implies smaller statistical variability for the covariance than a peaked Co-spectrum. In our estimates we have taken the observed shape of the CO₂ Co-spectrum into account when calculating the estimate of the statistical variability of the covariance, see e.g. Bendat and Piersol, Ch. 7 (2010).

Bendat, J.S. and Piersol, A.G.: Random Data; Analysis and Measurement Procedures. (4th ed.). John Wiley and Sons, New York, 2010.

Editor question: Is the following reason related to the shift in wind directions reason conveyed in the text for future readers?

“Fig 4. First 3 days 8/25-8/28 appear most ‘active’ in terms of the pollutant spikes or features in these time series in panels a,b,e. Then a precipitous drop at 8/28 around 1200 (precip?), after which fewer pollutant spikes.

We clarified the text on page 12, lines 341-353:

The 96 h backward trajectory analysis of Flextra by NILU (Stohl et al., 1995) showed that in the measurement period before the noon of 28 August an air mass was transported through central Europe and arrived in Helsinki (Fig. 5a) carrying anthropogenic pollutants. A sudden drop in the concentrations of N_{tot} and PM₁ occurred at noon on 28 August when the wind turned and blew from the west until 10 AM on 30 August (Fig. 4c) over the Atlantic and Baltic Sea carrying clean air with low particulate concentrations (Fig. 5b). Simultaneously, the diurnal variation of CO₂ diminished. During that period there was no precipitation. During the last 12 hours before the clean air mass arrived in Harmaja the average background particle concentrations stayed rather constant at $\sim 2.7 \times 10^3 \text{ \#/cm}^3$, whereas the PM₁ increased from $\sim 4 \text{ \mu g/m}^3$ to $\sim 11 \text{ \mu g/m}^3$. This indicates that also larger particles were transported from Europe. In fact, this is obvious from Fig. 6, which presents the average number size distribution (Fig. 6a) as well as the volume size distribution (Fig. 6b) of background particles in the evening of 27 August, at noon on 28 August just before the clean air mass arrived, in the afternoon of 28 August, and early in the morning of 1 September. In the last-mentioned case the particle number concentration was highest ($3.6 \times 10^3 \text{ \#/cm}^3$), but due to the small particle sizes (Fig. 6a) they did not have an effect on the volume (and mass) size distribution (Fig. 6b).

Note:

Because in the manuscript text all CO₂ fluxes by the EC method are given in the unit of $\mu\text{mol m}^{-2} \text{s}^{-1}$, we converted the median F_{CO_2} ($\text{mg m}^{-2} \text{s}^{-1}$) in Table 1 accordingly. We also found a typing error in its uncertainty value, which should be read 30.0%.