

acp-2018-603: Surface–atmosphere exchange of inorganic water-soluble gases and associated ions in bulk aerosol above agricultural grassland pre- and post-fertilisation

by Ramsay, R. et al.

Response to Anonymous Referee #4

We thank the reviewer for their time in reviewing our paper. The reviewer has provided an additional comment in their report, and we shall address their concern in this response.

“My only additional comments regards the concentration footprint: flux and concentration footprints are generally different, with the concentration ones having a wider area compared with the flux ones. Is the area around the sampling point homogeneous enough for discussing fluxes and concentrations "hand-in-hand" (also considering flux divergencies)?”

We struggle to fully understand the point the reviewer is making and consequently to address their concern. It is correct that concentration and flux footprints are very different, with the flux footprint extending to typically 100× the measurement height (depending on atmospheric stability) and the concentration footprint being much larger. We do discuss both concentrations and fluxes in the paper, but at no point state that the concentrations are only affected by the field itself. By contrast we did confirm through footprint estimation that the reported fluxes were dominated by the field itself. Otherwise a flux measurement by the aerodynamic gradient technique would indeed not be meaningful. For this assessment it is important to understand that for gradient flux measurements the flux footprint describes the footprint of the *concentration difference* between the two heights rather than the footprint of the concentration measured at any single height itself.

Flux divergence on the other hand is caused by the local interaction between vertical transport and chemistry, depending on the relative time-scales of the two processes.

Whilst the footprints are different, the exchange mechanisms studied above this particular field will obviously apply more widely and therefore affect regional concentrations. For example, the elevated deposition velocity for the coarse aerosol fraction will also apply to the wider landscape and result in a decreased lifetime and transport distance of this aerosol fraction during transport to the site.

In summary, we do not see any conflict in discussing both concentrations and fluxes in the present manuscript and cannot see where we may have confused the reviewer.

Response to Co-Editor

We thank the co-editor for overseeing the review process for this paper.

We have noted the co-editor’s concern over the length and coherence of the introduction, and we have therefore taken steps which resolve this. With regards to length, we have shortened the introduction by 231 words, from 1792 to 1561 words, by omitting supplementary information that was not essential for the overall study justification. We have also improved clarity by rearranging sections of the introduction. The justification for the study is presented in the first paragraph, followed by short introductions to the inorganic trace gases and associated aerosol counterparts measured, with a brief overview of the aerodynamic gradient method and instrumentation.