Twelve years of nitrogen deposition gap? An EMEP4UK model analysis

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Abstract
An analysis of 12 years of annual nitrogen and sulphur deposition over the UK was carried out comparing atmospheric chemistry transport model (ACTM) results with an observation-derived calculation (CBED). The two deposition estimates agree well for oxidised sulphur, whereas total oxidised nitrogen deposition was underestimated by the ACTM. Possible causes of this discrepancy are the uncertainties of emissions estimates and the simplification in the ACTM aerosols formation. The CBED deposition estimates are less sensitive to uncertainties in the emissions inventory dataset as the UK deposition values are derived from observed deposition and surface concentrations. However, CBED wet deposition may be over-estimated due to dry deposition to the surface of bulk collectors. The UK deposition estimates show a general decline over the 2000-2012 period investigated; for oxidised sulphur ~86 (ACTM) and ~97 (CBED) Gg S yr^{-1}, for oxidised nitrogen ~29 (ACTM) and ~45 (CBED) Gg N yr^{-1}, and for reduced nitrogen ~7 (ACTM) and ~5 (CBED) Gg N yr^{-1}.

Key Words
Nitrogen, deposition, transport, chemistry, atmospheric model

Introduction
In the UK the main components of nitrogen deposition are oxidized nitrogen (OXN), and reduced nitrogen (RDN). Across Europe emissions of oxidized nitrogen (NO_x) and sulfur dioxide (SO_2) are reported to have steadily decreased in the last decade, whereas ammonia (NH_3) emission estimates have remained relatively stable. The analysis presented here aims at assessing to what extent atmospheric chemistry transport models, based on official emission inventory datasets, can reproduce these trends compared to observations.

Methods
An atmospheric chemistry transport model (ACTM) and the UK observation-derived (CBED) sulphur oxides (SOX), OXN and RDN UK depositions were compared for the years 2001-2012. The ACTM used here was the EMEP4UK model (Vieno et al., 2014, 2016) driven by the Weather and Research Forecast model (WRF – www.wrf-model.org). EMEP4UK used emissions inventories based on the spatial distribution of the UK National Atmospheric Emission Inventory (NAEI) estimates for 2012 and rescaled to the UK national total for each year. The CBED procedure (Smith et al., 2000) used observations and interpolation of measured concentrations of sulphur and nitrogen compounds in air and precipitation across the UK to derive a spatially explicit UK deposition budget.

Results
Figure 1 shows the UK NAEI data for the year 2001-2012 for ammonia (NH_3), sulphur dioxide (SO_2) and nitrogen oxides (NO_x). This figure illustrates the substantial reduction of UK SO_2 and NO_x but relatively stable NH_3 emissions to the atmosphere during this period.
The UK reductions in SOX, OXN, and RDN depositions are presented in Fig. 2, 3, and 4, respectively, and show a general decline for both the ACTM and CBED estimates. For SOX a reduction of ~86 (ACTM) and ~97 (CBED) Gg S yr^{-1}, for OXN a reduction of ~29 (ACTM) and ~45 (CBED) Gg N yr^{-1}, and for RDN a reduction of ~7 (ACTM) and ~5 (CBED) Gg N yr^{-1}. However, the ACTM underestimates the absolute value especially for OXN and RDN deposition. The discrepancy between the ACTM and CBED estimate may be explained by the underestimation of UK NO_x emissions. However, as dry deposition values are similar between both approaches, compared with wet deposition, especially for OXN and RDN, this may suggest that mainland European emissions uncertainties are more likely the cause. Also, the CBED wet deposition may be over-estimated due to dry deposition to the surface of bulk collectors (Cape et., al. 2009).

A reduction of ~60% UK SO_2 emissions (Figure 1) is associated with a reduction of 53% (ACTM) and 46% (CBED) in SOX deposition, a reduction of 39% UK NO_x emissions is associated with a 28% (ACTM) and 28% (CBED) reduction in OXN deposition, and a reduction of ~14% of UK NH_3 emission is associated with

a 5% (ACTM) and 3% (CBED) reduction in RDN deposition. European emissions reductions between 2001 and 2012 will have also impacted on UK deposition, as between 50% and 60% of secondary inorganic aerosols are directly imported from mainland Europe and contain N and S in the form of ammonium nitrate and sulphates (Vieno et. al., 2016).

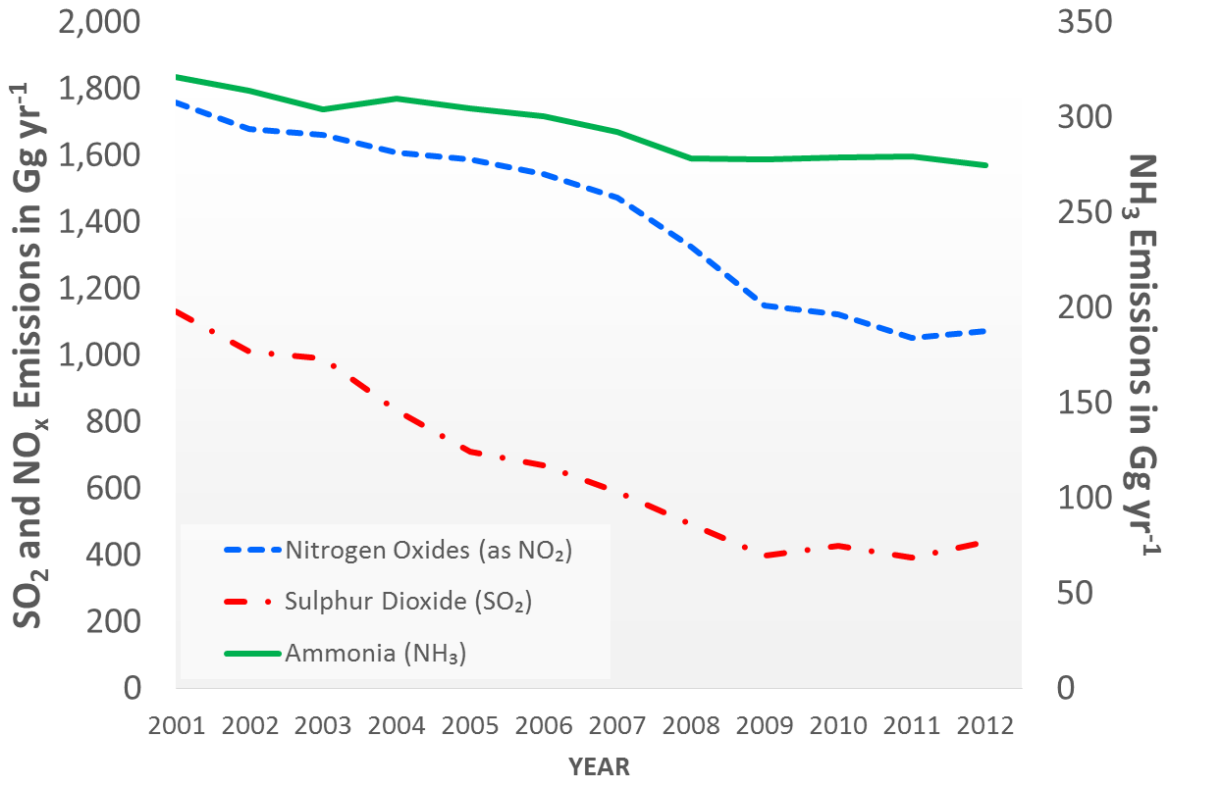

Figure 1. UK National Atmospheric Emission Inventory (NAEI) data for the years 2001-2012 (http://naei.defra.gov.uk/).

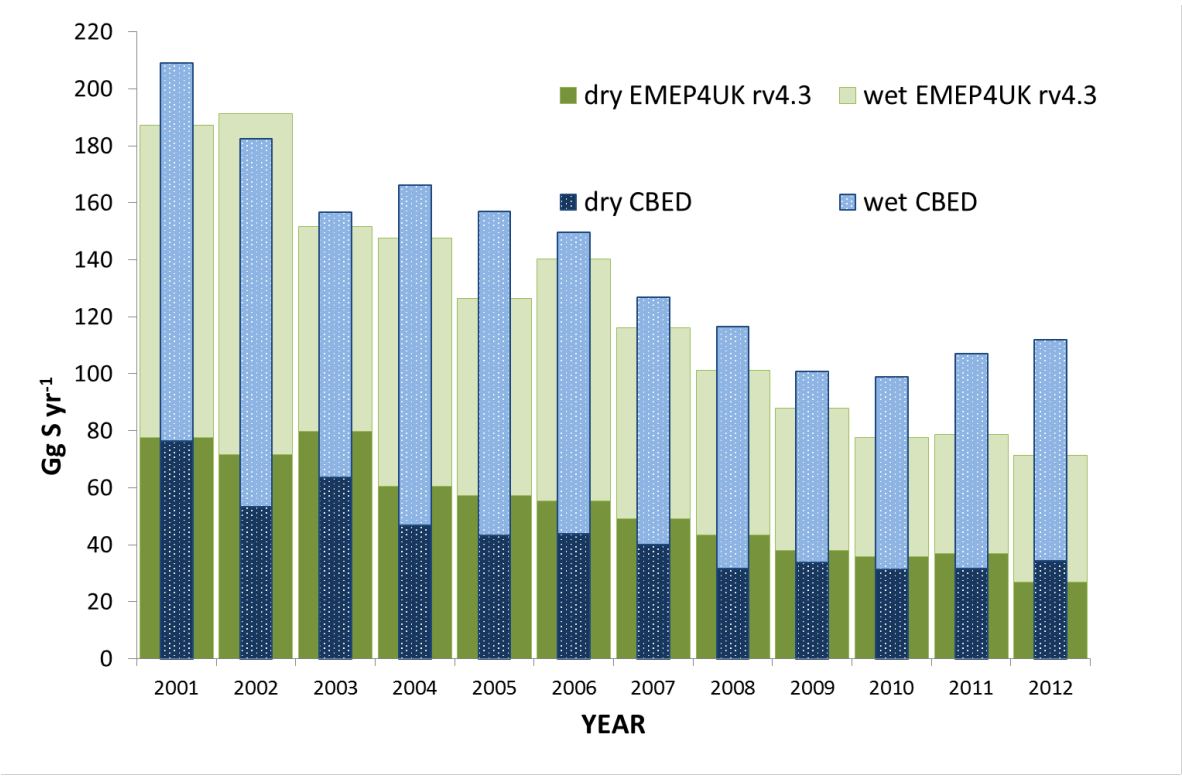

Figure 2. EMEP4UK dry (solid dark green) and wet (solid light green) deposition of SOX, and from the CBED observation derived dry (dark blue) and wet (light blue) deposition for the years 2001-2012.

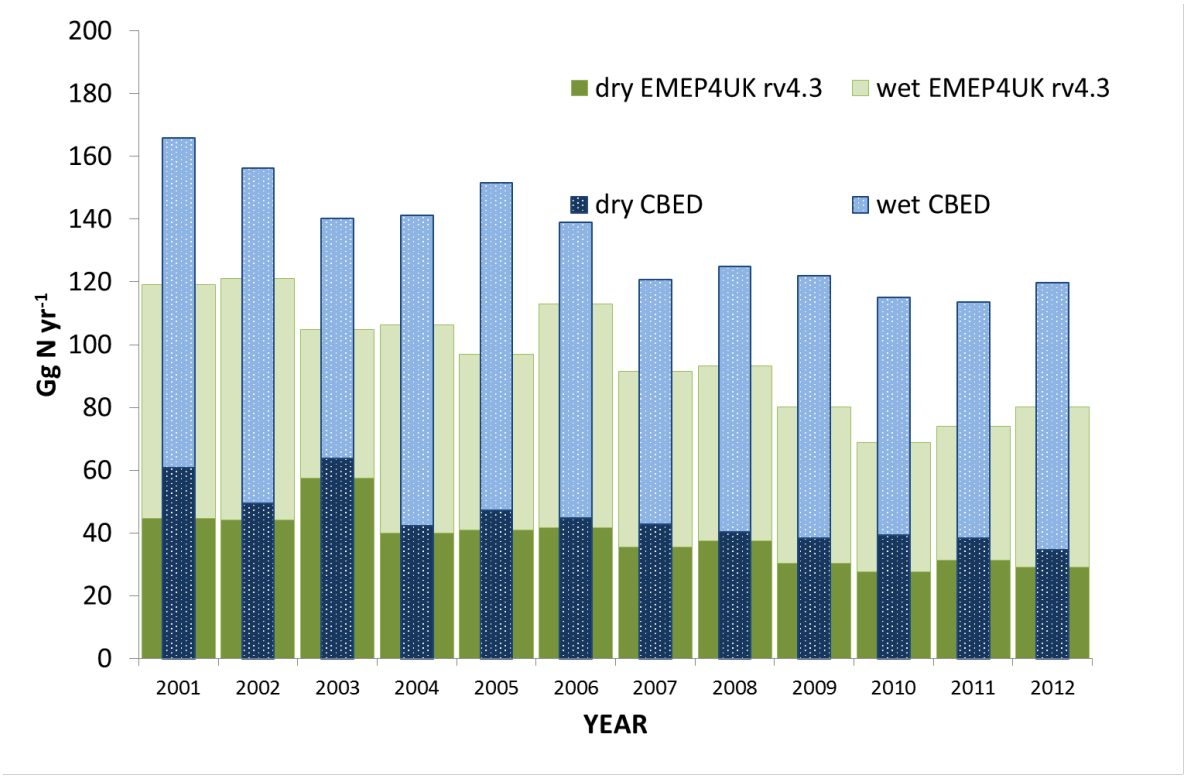

Figure 3. EMEP4UK dry (solid dark green) and wet (solid light green) deposition of OXN and from the CBED observation derived dry (dark blue) and wet (light blue) deposition for the years 2001-2012.

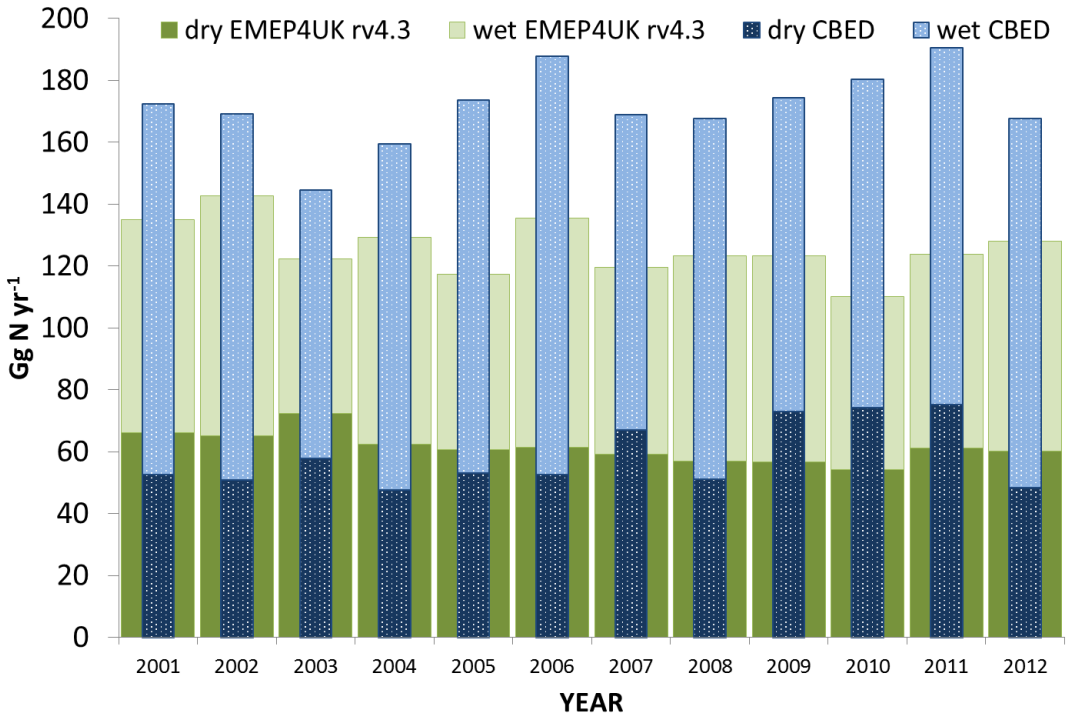

Figure 4. EMEP4UK dry (solid dark green) and wet (solid light green) deposition of RDN, and form the CBED observation derived dry (dark blue) and wet (light blue) deposition for the years 2001-2012.

Conclusion

The main conclusion is that uncertainties in emissions estimates may contribute to the discrepancy between the CBED and ACTM (EMEP4UK) estimates. However, the deposition gap may also be explained by the shortcoming of the ACTM itself, or overestimations of values in observations used in the CBED approach. Although total deposition is underestimated by the ACTM compared with CEBD, the dry deposition fraction is generally well modelled by the ACTM, whereas wet deposition is underestimated. Wet deposition is

associated with long-range transport whereas dry deposition is more closely linked with local emissions, suggesting that emissions contributions from sources elsewhere in Europe, or the extra-European boundary conditions, are underestimated. This has not been formally tested and further work is required for a proper assessment. However, this initial study shows how model results and observations can be combined to support a more comprehensive assessment of the underlying drivers of trends in atmospheric concentrations and depositions (Reis et al., 2016).

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