Replies to the Anonymous Referee 1

We thank the referee for the valuable comments which helped us to improve the manuscript. Please find below our responses (in black) after the referee comments (in blue). Changes in the revised manuscript are written in *italics.*

This MS presents a modelling study dealing with the role of ammonia on air quality in Europe, with a focus on shipping as a key emission sector. The text is straight forward and well-written, and of interest to the scientific community. I have only minor issues which should be clarified prior to publication:

- line 51: what is the reason behind the increase in ammonia emissions since 2014?

Agriculture is the main source of ammonia and emissions mainly result from the stabling of animals and the storage and application of animal manure. The application of inorganic N-fertilizers is also a source of ammonia emissions. Emissions decreased between 1990 and 2000 in Europe mainly due to declining numbers of animals. After 2000, the decrease in European countries slowed and emissions even started to increase, especially in the eastern part of Europe. The increase in ammonia emissions since 2014 is due to the difficulty in implementing further emission reductions, especially in the agriculture sector. We added the following sentence in the revised manuscript:

P3, L56

Ammonia emissions have been increasing again since 2014, however, posing problems for Europe (NEC, 2019). This is mainly due to the difficulty in implementing additional emission reductions in the agriculture sector, especially in the housing of animals and the storage and application of animal manures.

- line 67: what is the status of this implementation? Have these new sulfur emission regulations been effectively implemented (as they were supposed to start in 2020)?

The new regulation has been in force since 1 January 2020. It reduces the limit for sulfur in fuel oil used in ships operating outside designated emission control areas to 0.50%. IMO reports that it worked with the Member States as well as the shipping and refining industries to identify and mitigate transitional issues so that ships meet the new requirements https://www.imo.org/en/MediaCentre/PressBriefings/Pages/34-IMO-2020-sulphur-limit-.aspx). There are also guidelines being developed by IMO for consistent implementation of the MARPOL regulation coming into effect from 1 January 2020. Monitoring, compliance and enforcement of the new limit is the responsibility of governments and national authorities of Member States that are parties to MARPOL Annex VI. Flag States (the State of registry of a ship) and Port States also have rights and responsibilities to enforce compliance.

- line 77: same as above, what is the status of this statement? "According to the European Environment Agency, emissions of nitrogen oxides from international maritime transport in European waters are projected to increase and could be equal to land-based sources by 2020 (EEA, 2013). " The reference dates back to 2013, do the authors have data on the current emissions? How accurate was EEA's projection from2013?

To our knowledge, there is as yet no current update on the land versus ship emissions in 2020 (this year). More recent projections with current emission control regulations, however, indicate that NOx emissions from international shipping will exceed those from land-based sources in the EU after 2030 (Cofala, 2018, http://pure.iiasa.ac.at/id/eprint/15729/).

- line 120: the scenarios "current legislation (CLE)" refer to the regulations included the lower sulfur limits from 2020 (see comment to line 67)? Or prior to 2020? Please clarify.

CLE 2020 refers to emissions according to the regulations in current legislation with the limits coming into force at the beginning of 2020.

- line 135, please review sentence (2 verbs): "The model results for 1990, 2000 and2010 were compared with the measurements available at the EDT project database based on EMEP datasets and model performance for SO2, NO2, PM10, PM2.5 and hourly O3 was discussed in detail in Jiang et al. (2020)."

We rephrased the sentence as follows:

P6, L179

The model results for 1990, 2000 and 2010 were compared with the measurements available at the EDT project database which is based on EMEP datasets (https://wiki.met.no/emep/emep-experts/tfmmtrendstations). The number of available measurement stations varies between 15 and 64 depending on the year and species. For ozone, only measurements at the background-rural stations were used to reduce uncertainties due to the model resolution. Model performance for SO_2, NO_2, PM_{10}, $PM_{2.5}$ and hourly O_3 was discussed in detail in Jiang et al. (2020).

- line 140: measuring ammonia is rather complex, therefore the quality of the observations should also be discussed (even if briefly). Please add some information on the measurement method and comparability between (the few) ammonia datasets available.

We provided detailed information about the measurements in a new table in the Supplementary (Table S1) and added the following statements in Section 3.1.:

P6, L188

Atmospheric concentrations of ammonia are not well characterized due to relatively small number of monitoring sites, the short lifetime of NH_3 in the air and the difficulty of measuring non-point source emissions such as agricultural fields. Most of the measurement sites used in this study are located in the north; only very few stations are in the other parts of Europe (Fig. 1). The detailed information about the measurements (location, methods, temporal resolution) at each site is given in Table S1. Most of the measurements are daily concentrations, except for some sites in the Netherlands (hourly), Spain and Italy (weekly), Switzerland (bi-weekly) and UK (monthly). Measurement methods also differ; most of the stations use filter-pack sampling, while the passive samplers were used at 2 sites in Spain and the denuder systems were adopted at sites in the Netherlands, Great Britain and Switzerland. One should keep in mind that sampling artefacts due to the volatile nature of ammonium nitrate and the possible interaction with strong acids make separation of gases and particles by simple aerosol filters less reliable as indicated by EMEP (Co-operative Programme for Monitoring and Evaluation of the Long-Range Transmission of the Air

Pollutants in Europe), (https://projects.nilu.no/ccc/reports/cccr1-2019_Data_Report_2017.pdf). Modelled ammonia concentrations are similar to the measured ones at the few sites in the south while one site in eastern Europe shows an underestimation (Fig. 1). On the other hand, ammonia is overestimated at several sites such as in the UK, and in high emission areas around the Netherlands and Denmark. The mean fractional bias at all sites is 37.9% (Table S2).

- line 156: "and/or deposition is underestimated by the model for which the resolution might also be critical factor". Is the model resolution not the same for all regions? If it is, then it could not explain the differences between central Europe and Iberian Peninsula and in Scandinavia (Fig. 1). Can the authors provide an explanation about why deposition might be more underestimated in central Europe than in the Iberian Peninsula and in Scandinavia? This seems a more likely cause for the model's overestimation around these high emission areas.

The horizontal resolution is the same everywhere in the model domain. The model performance for deposition, however, might differ in different regions in the domain due to various factors:
The spatial resolution used for the model simulations can add uncertainty to the model results, since the model estimate for a grid cell might not be representative of the specific location of the measurement site. Especially in mountainous areas with high spatial variability in precipitation patterns, spatial representativeness of the measurement sites is not possible. Furthermore, measurement sites close to farming areas may overestimate deposition of reduced nitrogen with respect to the modelled average deposition within the grid cell. Central Europe has more agricultural area and cattle farming than Scandinavia and the Iberian Peninsula. In addition, several studies show that the dry deposition velocity of ammonia (which is calculated using turbulent diffusion and surface characteristics in models) might vary significantly depending on the season and region (Flechard et al., 2011; Aksoyoglu and Prévôt, 2018). Different regional performance of the parameters used in the calculations might lead to different model performance for deposition. As reported by Theobald et al. (2019), the tendency of models to underestimate wet deposition and overestimate atmospheric concentrations is likely due to deficiencies in simulating wet-deposition processes, which are related to the vertical concentration profiles, scavenging coefficients or in-cloud processes, including the parameterisation of clouds.

We added the following paragraph in the revised text:

P7, L216
These results suggest that ammonia emissions in the emission inventory might be too high around the main emission sources in central Europe and/or deposition is underestimated by the model for which the resolution might also be a critical factor. The model estimate for a grid cell might not be representative of the specific location of the measurement site. Especially in mountainous areas with very spatially variable precipitation patterns, spatial representativeness of the measurement sites is not possible. Furthermore, measurement sites close to farming areas may overestimate deposition of reduced nitrogen with respect to the modelled average deposition within the grid cell. In addition, several studies show that the dry deposition velocity of ammonia (which is calculated using turbulent diffusion and surface characteristics in

models) might vary significantly depending on the season and region (Flechard et al., 2011; Aksoyoglu and Prévôt, 2018). Therefore, different regional parameters used in the calculations might lead to different model performance for deposition. Moreover, as reported by Theobald et al. (2019), the tendency of models to underestimate wet deposition and overestimate atmospheric concentrations is likely due to deficiencies in simulating wet-deposition processes, which are related to the vertical concentration profiles, scavenging coefficients or in-cloud processes, including the parameterization of clouds.

- line 171: "Among the SIA components, the best agreement between model and measurements is for sulfate". Can the authors quantify the relative difference (in %) between modelled and measured concentrations, for sulfate and nitrate respectively? It would be useful for the reader to have this information as well as the absolute difference (in microg/m3 shown in the Figure). Also for ammonia, in the previous paragraphs.

The relative difference between modelled and measured concentrations can be seen in Table S2 in the Supplement. For instance, mean fraction bias for sulfate is 4.7% while for nitrate it is 54.6%. For ammonia the MFB is 37.9%. We added this information from the supplement to the revised text as follows:

P7, L201

On the other hand, ammonia is overestimated at several sites such as in the UK, and in high emission areas around the Netherlands and Denmark. The mean fractional bias at all sites is 37.9% (Table S2).

P8, L245

Among the SIA components, the best agreement between model and measurements is for sulfate (MFB = 4.7%) (Table S2, Fig. 1). The modeled concentrations of the other SIA components - for which the spatial coverage in central and western Europe is rather poor - are higher than the measured ones, especially for nitrate (MFB = 54.6%) (Fig. 1, Table S2).

- lines 180-185: similarly to above, what is the reason for the increases in ammonia emissions? Is it increases in key sources (agriculture)? Or to a mix of emissions and atmospheric processes?

Please see the explanation above.

- line 197: "On the other hand, since simulations for 2030 were performed using the meteorological parameters of 2010, one should keep in mind that potentially higher temperatures in the future might increase the evaporation of ammonium nitrate to form its gaseous components NH3 and HNO3". This is a key point which could be high-lighted in the abstract.

Thank you for this suggestion. We added the following sentence in the abstract:

P2, L29

One should also keep in mind that potentially higher temperatures in the future might increase the evaporation of ammonium nitrate to form its gaseous components NH_3 and HNO_3.

- line 217: as above, please clarify what is meant by "current legislation" (before or after 2020): "Results of future scenario simulations suggest that sulfate concentrations will continue to decrease in central Europe as well as along shipping routes until 2030 assuming a current legislation (CLE) scenario (Fig. 3d, right panel)"

Current Legislation (CLE) is defined as legal and regulatory provisions in place at a certain agreed date. The ship emissions in 2020 and 2030 are projected based on current legislation (CLE) of the International Maritime Organization (IMO) and the EU. In such emission scenarios, it is assumed that emissions will be reduced by the amounts defined for 2020 and 2030 with respect to the reference year 2005.

- section 3.4: only as a suggestion, it might have been interesting to add an additional scenario including the implementation of a SECA in the Mediterranean (Rouïl,L., Ratsivalaka, C., André, J.-M., Allemand, N., 2019. ECAMED: a Technical Feasibility Study for the Implementation of an Emission Control Area (ECA) in the Mediterranean Sea. IMO report MEPC 74/INF.5.). An analysis of the potential impacts/benefits of this potential SECA in the framework of the authors' study could be very useful.

This is a very good suggestion. For a proper scenario calculation, however, the emission inventory has to be regenerated with modified ship emissions based on the SECA requirements. This can be done in a future project, but unfortunately not for this present study.
Moreover, similar scenario simulations have already been done. For example, Cofala et al., (2018) reported that designation of the Mediterranean Sea as an ECA could by 2030 cut emissions of SO_2 and NO_x from international shipping by 80 and 20 percent, respectively, compared to current legislation.

- Table 1: please define the acronyms (CLE, MTFR) in the table header.
Done.

Aksoyoglu, S., and Prévôt, A. S. H.: Modelling nitrogen deposition: dry deposition velocities on various land-use types in Switzerland, Int. J. Environ. Pollut., Vol. 64, 230–245, https://doi.org/10.1504/IJEP.2018.10020573, 2018.

Flechard, C. R., Nemitz, E., Smith, R. I., Fowler, D., Vermeulen, A. T., Bleeker, A., Erisman, J. W., Simpson, D., Zhang, L., Tang, Y. S., and Sutton, M. A.: Dry deposition of reactive nitrogen to European ecosystems: a comparison of inferential models across the NitroEurope network, Atmos. Chem. Phys., 11 2703-2728, 10.5194/acp-11-2703-2011, 2011.