Atmos. Chem. Phys. Discuss.,
https://doi.org/10.5194/acp-2017-1184-AC1, 2018

Atmospheric Chemistry and Physics Discussions

Interactive comment on "High resolution modeling of gaseous methylamines over a polluted region in China: Source-dependent emissions and implications to spatial variations" *by* Jingbo Mao et al.

Jingbo Mao et al.

maojingbo03@163.com

Received and published: 14 April 2018

We would like to thank the referee for his/her constructive comments which help to improve our manuscript. Our point-to-point replies (in blue) to the comments are given below (the original comments are copied here in black). The manuscript has been revised accordingly. All the changes to the manuscript have been highlighted using the Microsoft word "track-changes" tool in one version of the submitted revised manuscript.

Anonymous Referee 1

Gaseous amines represent a category of base compounds which plays import roles in many aspects of atmospheric chemistry including nucleation and growth of newly formed particles. Compared to ammonia, concentrations of individual amines are several orders of magnitude lower, far below ppb levels. In addition, there are a variety of sources of amines in the atmosphere. Furthermore, most amines are rather reactive, bearing shorter lifetimes than ammonia. Hence the temporal and spatial distributions of amines can vary significantly. This paper presents a high resolution modeling study of methylamines (C1-C3) in Yangtze River Delta Region (YRD) by considering source dependent amine-to-ammonia ratios (SDR) whose results demonstrate much better agreement with observations than those assuming fixed ratios (FR) in the model simulations. Here four domains are considered and the simulated results from the smallest two domains showed that models with higher spatial resolution yield better agreement with observations, demonstrating the need for employing high resolutions when modeling spatial distributions of amines in order to better understand their roles in atmospheric chemistry.

Thanks for the nice summary and the positive comments.

The paper can be publishable after the following issues are resolved:

1. The paper models the amine concentrations and their spatial distributions from five different source types (chemical industry, other industry, agriculture, residential, and transportation). What is the rationale behind this classification? Are there any previous studies that employed a similar classification?

In various emission inventories such as MEIC and INTEX-B, emission sources are generally separated into different types like residential, agriculture, transportation, industry, and power plant. Considering that emission rates of amines from organic synthesis may differ significantly with those from power generation and heavy industries using selective catalytic reduction (Zheng et al., 2015), we divided industrial sources into chemical industry and other industry in the present study. As emphasized in the Intro-

duction, no previous modeling studies (to our knowledge) have distinguished different amines emission from various source types.

2. The study used measured data from two sites (NUIST and Fudan sites). Since the measured amine concentrations might be strongly affected by the close-to-site sources, the authors should provide some evidences that those sites are not significantly affected by local sources which may lead to systematic biases for the data. According to Table 4, the Fudan site may be affected significantly by local sources.

In this study we used amines to ammonia ratios in various plumes observed at the NUIST site to derive source-dependent amines emissions. Ammonia was not measured at the Fudan site during the period when amines were measured. We agree that concentrations of amines can be strongly affected by the close-to-site sources. Nevertheless, we do not use absolute concentration of amines, but the ratios of amines to ammonia to derive amine emissions. Therefore, the effect of local sources does not impact the conclusions of this paper.

3. Table 3 lists emission rates of C1-C3 amines from different sources based on the SDR ratios from this study. However, it is not very clear how those values are obtained. In section 2.2, the authors only used SDR from the data measured in 2012 (NUIST site) and did not even mentioned those measured in 2015 (Fudan site). The authors should provide the reasons for only considering one data set rather than both data sets. In addition, the paper mentioned very briefly the uncertainties associated with the measured data. Can those uncertainties be quantified? How a single (or even two) measured site can be representative of the domains of interest (i.e., D3 and D4)? How those five different sources of amines are determined, for example, based on what criteria, the emission rates of the five sources are distributed?

We derived emission rates of C1-C3 amines listed in Table 3 based on SDR rations and ammonia emissions from different sources. We have refined relevant description in the manuscript to make this clearer.

We only used SDR from the data measured in 2012 (NUIST site) because only the NUIST data has simultaneous measurements of NH3, NOx, and SO2 along with amines, enabling us to identify the plume source types (as detailed in Zheng et al., 2015). These species were not measured at the Fudan site during the period when amines were measured.

As emphasized in the manuscript, the estimation of amines emissions from different sources is subject to a large uncertainty, mainly due to very limited measurements available to constrain the estimation. We agree with the reviewer's concern about the representativeness of limited measurements and this can only be resolved by more similar measurements. The present study is the first attempt (to our knowledge) to use direct measurements to constrain amine emissions from different sources. The SDR approach, as we show here, improves the skill of the model in simulating concentrations of amines in polluted regions. We hope more field observations as well as more accurate source apportionment of amines will be carried out in the future to constrain the amines emissions and model study.

We derived regional methylamines emissions based on amines to ammonia ratios and ammonia emissions from different sources. The temporal and spatial distributions of C1-, C2-, and C3-amines follow those of ammonia for different sources.

4. Some rather minor points:

1) L7 on p2, change "model's" to "of the model"; similarly for "model's skill" on p7 (L27)

Modified.

2) L27 on p4, change "amines concentrations" to "concentrations of amines"; there are lots of those usages throughout the paper. Please correct them;

Modified.

3) L1-2 on p5, year 2014 is not up-to-date;

We mean the emission inventory for the year 2014 is up-to-date. To avoid confusion, we have deleted “up-to-date” from the sentence.

4) L23 on p5, C2 change “The point sources data” to “the data of the point sources”;

Modified.

5) L15 on p6, “at an urban site” not “in an urban site”;

Modified.

6) L21 on p6, delete “seek to”;

Modified.

7) L9 on p7, “in details” not “in detail”;

Modified.

8) L15-20 on p7, this ratio of 0.026 might be problematic if the measured site is so close to the source and affected strongly by the emissions from the source;

The ratio of 0.026 is detected in the ammonia water from a local chemical supplier.

9) L23 on p7, delete “would like to”;

Modified.

10) L4 on p8, “prior to this study” might be better replaced by “in previous studies”;

Modified.

11) L9 on p10, change “that” to “those” since it refers to as “distributions”;

Modified.

12) L28 on p10, “general underprediction of the model”, do you mean that it is compared to measurements?

Yes.

13) L10-11 on p11, where those values are from?

We derived the values based on Fig.5-6 and Table 4.

14) L18-20 on p11, I don’t think wind direction and speed are the reasons.

It is noted that many reasons may cause the underestimation, but at least partially due to the large deviation of the simulated wind directions and speeds during the period.

Please also note the supplement to this comment:
https://www.atmos-chem-phys-discuss.net/acp-2017-1184/acp-2017-1184-AC1-supplement.pdf

---

Interactive comment on Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2017-1184, 2018.