Towards an experiment to investigate N_2O_5 uptake to aerosol particles at ambient conditions using the radioactive tracer ^{13}N

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INTRODUCTION

N_2O_5 is an important atmospheric trace gas that plays a significant role in **nighttime tropospheric chemistry** [1]. The importance of N_2O_5 stems from its role as a **NO_3 radical reservoir** and a **major sink for NO_x species** thanks to heterogeneous loss processes. This leads to a reduction of tropospheric ozone and lowering of the oxidizing capacity of the troposphere [1, 2]. Laboratory studies have also shown that uptake of N_2O_5 to aerosol particles depends on meteorological parameters like temperature and relative humidity as well as aerosol composition [1, 2].

EXPERIMENTAL

In our experiments the short-lived radioactive tracer ^{13}N technique (PROTRAC) [3] developed at the Paul Scherrer Institute was coupled to an aerosol flow tube reactor. This method allows for the study of N_2O_5 uptake kinetics under realistic conditions but should also allow to observe behavior in a **wide temperature range** (tropospheric conditions) and the **influence of concentration effects** (nitrate effect).

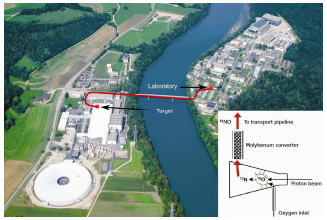

Fig 1: PROTRAC facility at PSI

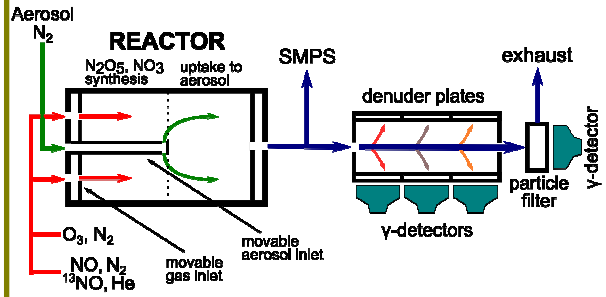

Fig 2: Schematic representation of the experimental setup

^{13}NO produced at the PROTRAC facility is mixed with non-labeled NO and O_3 in the reactor, where N_2O_5 is synthesized. The ^{13}N labeled species formed are trapped inside a narrow **parallel plate diffusion denuder system**, which allows for a selective separation of the gaseous species present. The aerosol particles are trapped on a **particle filter** located at the exit of the denuder system. Activity of the ^{13}N labeled species deposited on the denuder plates and particle filter is monitored via **scintillation counters**.

FIRST RESULTS

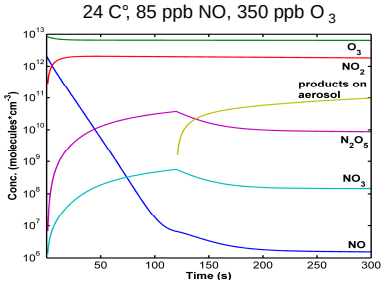

Fig 3: Example of modeling output

In order to optimize N_2O_5 production, study concentration and temperature dependence and evaluate potential problems that might arise (eg. N_2O_5 formation time vs. half-life of ^{13}N), N_2O_5 gas-phase formation was modeled in Matlab.

Several denuder coatings for N_2O_5 were tested and **citric acid** was selected because of its lower interference with NO_2.

A comparison of modeled vs. measured N_2O_5/NO_2 ratios has shown that the values are of the same order of magnitude. The differences can be attributed to radioactive decay and larger then predicted wall loss.

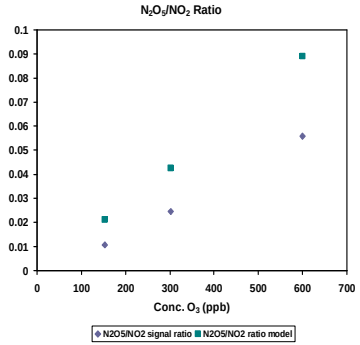

Fig 4: Modeled and measured N_2O_5/NO_2 ratios

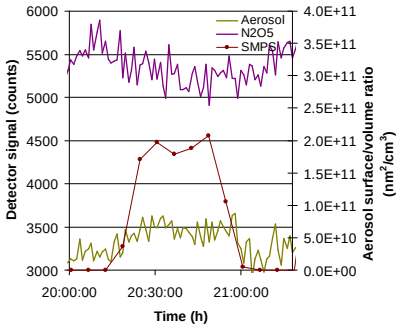

Fig 5: N_2O_5 uptake on citric acid aerosol

A **preliminary study** using a **citric acid aerosol** at relative humidities of 50-80% RH has shown **uptake of radioactively labeled N_2O_5** on the aerosol particles. First results obtained from a limited data set show an **uptake coefficient** γ in the order of **10^{-3}**.

SUMMARY

- ^{13}N labeled N_2O_5 has been produced for the first time using ^{13}NO produced at the PROTRAC facility at PSI
- Gas-phase formation of N_2O_5 in the flow reactor has been successfully modeled in Matlab and the model has shown good accordance with experimental observations
- Uptake of ^{13}N labeled N_2O_5 on citric acid aerosol has been observed

REFERENCES

[1] Chang, W. L., et al., Aerosol Sci. Technol., 45, 665-695 (2011).
[2] Finlayson-Pitts, B. J., and J. N. Pitts, Jr., Academic Press, San Diego, CA (2000).
[3] Ammann, M., Radiochim. Acta, 89, 831-838 (2001).

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