Scanning Infrared Remote Sensing System for Identification, Visualization, and Quantification of Airborne Pollutants

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ABSTRACT
Remote sensing by Fourier-transform infrared (FTIR) spectrometry allows detection, identification, and quantification of airborne pollutants. In the case of leaks in pipelines or leaks in chemical plants, chemical accidents, terrorism, or war, hazardous compounds are often released into the atmosphere. Various Fourier-transform infrared spectrometers have been developed for the remote detection and identification of hazardous clouds. However, for the localization of a leak and a complete assessment of the situation in the case of the release of a hazardous cloud, information about the position and the size of a cloud is essential. Therefore, an imaging passive remote sensing system comprised of an interferometer (Bruker OPAG 22), a data acquisition, processing, and control system with a digital signal processor (FTIR DSP), an azimuth-elevation-scanning mirror, a video system with a DSP, and a personal computer has been developed. The FTIR DSP system controls the scanning mirror, collects the interferograms, and performs the Fourier transformation. The spectra are transferred to a personal computer and analyzed by a real-time identification algorithm that does not require background spectra for the analysis. The results are visualized by a video image, overlaid by false color images. For each target compound of a spectral library, images of the coefficient of correlation, the signal to noise ratio, the brightness temperature of the background, the difference between the temperature of the ambient air and the brightness temperature of the background, and the noise equivalent column density are produced. The column densities of all directions in which a target compound has been identified may be retrieved by a nonlinear least squares fitting algorithm and an additional false color image is displayed. The system has a high selectivity, low noise equivalent spectral radiance, and it allows identification, visualization, and quantification of pollutant clouds.

Keywords: remote sensing; scanning FTIR spectrometer; toxic cloud imaging; visualization; quantification

1. INTRODUCTION
In the case of chemical accidents, leaks in pipelines, terrorism, or war, hazardous compounds are often released into the atmosphere. In order to take appropriate measures to protect workers, residents, emergency response personnel at the site of the release, and the environment, information about the released compounds is required immediately. Various analytical methods including mobile gas chromatography/mass spectrometry (GC/MS), infrared spectrometry, and hand-held sensors like photoionization detectors and electrochemical cells are applied. Recently, a portable gas detector array comprised of an ion mobility spectrometer in combination with a photoionization detector, an electrochemical cell and two semiconductor gas sensors has been developed for on-site analysis. However, all these methods require the collection of a sample, which may be dangerous. Passive remote sensing by infrared spectrometry allows detection and identification of hazardous clouds from a distance. For a complete assessment of the situation, information about the position and the size of the cloud is essential. Various imaging spectrometers that provide this information have been developed: systems with high spatial resolution based on forward looking IR systems combined with tunable filters and imaging Fourier-transform infrared (FTIR) spectrometers that use a detector array instead of a single detector. However, the noise equivalent spectral radiance (NESR) of these imaging spectrometers is greater than the NESR of remote sensing FTIR spectrometers with a single detector element. Various scanning FTIR systems, optimized for different applications have been developed. They provide high-quality spectral information (regarding the respective application) in combination with spatial information but lower temporal resolution. The scanning remote sensing system presented in this work provides a low limit of detection (low NESR), high selectivity (spectral resolution of 4 cm⁻¹), quantitative analysis, and visual information about the size and position of possibly hazardous clouds. The scanning system allows monitoring over wide geographic areas including regions that may be inaccessible.

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otherwise. The applied detection algorithm is described in detail in reference 7. This work focuses on a new quantification algorithm and presents the extension of the system by a video processing system.

2. IDENTIFICATION AND QUANTIFICATION OF AIRBORNE POLLUTANTS BY INFRARED REMOTE SENSING

2.1 Radiative Transfer Model

Passive remote sensing of hazardous clouds is based on the analysis of infrared radiation absorbed and emitted by the molecules of the clouds. Figure 1 illustrates the measurement setup of the method. The radiation measured by the spectrometer contains the spectral signatures of the background of the field of view, the vapor cloud, and the atmosphere. The propagation of radiation through the atmosphere is described by the theory of radiative transfer. In order to describe the basic characteristics of spectra measured by a passive infrared spectrometer a simple model with three layers can be used (Figure 1). Radiation from the background, for example, the sky or a surface (Layer 3) propagates through the vapor cloud (Layer 2) and the atmosphere between the cloud and the spectrometer (Layer 1). The layers 1 and 2 are considered homogeneous with regard to all physical and chemical properties. The radiation containing the signatures of all layers is measured by the spectrometer.

\[ L_1 = \left(1 - \tau_1\right)B_1 + \tau_1 \left(1 - \tau_2\right)B_2 + \tau_2 L_3, \]  

where \( \tau_i \) is the transmittance of layer \( i \), \( B_i \) is the spectral radiance of a blackbody at the temperature of layer \( i \), \( T_i \). \( L_i \) is the radiance that enters the layer of the cloud from the background. All quantities in Equation (1) are frequency dependent. If the background of the field of view is a surface, the radiation entering the cloud contains radiation emitted by the surface and reflected radiation, i.e. ambient radiation \( (L_{am}) \) and radiation from the sky \( (L_{sky}) \).

\[ L_3 = a \int_{\Omega_{s,0}}^{\Omega_{s,1}} F(\Omega_{s,0}, -\Omega') L_{sky}( -\Omega') d\Omega' + a \int_{2\pi - \Omega_{s,1}}^{\Omega_{s,0}} F(\Omega_{s,0}, -\Omega') L_{am}(-\Omega') d\Omega' + e(\Omega_{s,0}) B(T_{bg}) \]  

Here, \( a \) is the surface albedo, \( \Omega_s \) is the solid angle subtended by the aperture of the spectrometer. \( F(\Omega_{s,0}, -\Omega') \) is the surface biconical reflectance function for incident solid angle \( -\Omega' \) and emergent solid angle \( \Omega' \). The limits of the integrals are given by the total solid angle subtended by the sky \( \Omega_{sky} \). \( e(\Omega_{s,0}) \) is the directional surface emittance, and \( B(T_{bg}) \) is the radiance emitted by a blackbody at temperature \( T_{bg} \). The dependence on frequency is left implicit again. The
contribution of scattering is neglected. If the distance between the background surface and the cloud is long, i.e. the transmittance of the atmosphere is not negligible, another layer may be added.

If the temperatures of the layers 1 and 2 are equal, Equation (1) can be simplified:

$$L_1 = B_1 + \tau_1 \tau_2 (L_3 - B_1)$$  \hspace{1cm} (3)

It follows from Eq. (3) that in this case the layers 1 and 2 may be exchanged without any influence on the spectrum. The spectrum is independent of the position of the cloud between the background and the spectrometer. If the layers are exchanged, Eq. (1) becomes ($B_1 = B_2$)

$$L_1 = (1 - \tau_2)B_2 + \tau_2 B_1 + \tau_1 L_3.$$  \hspace{1cm} (4)

The term in square brackets is the background radiance $L_b$, i.e. the radiance without the presence of the cloud.

### 2.2 Identification

The identification method applied is based on the assumption that it is possible to approximate a spectrum with a linear combination of a limited number of reference spectra. First, the spectrum of the brightness temperature $T_{br}(\sigma)$ ($\sigma$: wavenumber) is calculated. The brightness temperature is defined by the Planck function $B$:

$$B(\sigma, T_{br}(\sigma)) = L(\sigma),$$  \hspace{1cm} (5)

where $L(\sigma)$ is the spectral radiance. It is calculated by solving Eq. (5) for $T_{br}$

$$T_{br}(\sigma) = \frac{h c \sigma}{\ln \left( \frac{2 h c^2 \sigma^3}{L(\sigma)} + 1 \right) + k}$$  \hspace{1cm} (6)

[$h$: Planck’s constant, $c$: speed of light, $k$: Boltzmann’s constant]. The emittance $\epsilon(\sigma)$ of many surfaces is high and almost constant in the range 650 – 1500 cm$^{-1}$. Thus, the emission spectrum of these materials has a high degree of similarity to the spectrum of a blackbody and the spectrum of the brightness temperature $T_{br}(\sigma)$ of these surfaces is almost constant. This makes the brightness temperature spectrum better suited for direct analysis than the radiance spectrum.

The spectrum is analyzed sequentially for all target compounds with different reference matrices. Each matrix contains spectral data of one target compound, atmospheric gases (H$_2$O, O$_3$), and the signatures of potential interferents. Moreover, it contains broad Gauss functions for the approximation of the baseline. The frequency dependence of the brightness temperature of the background and thermal effects inside the spectrometer may cause baseline shifts that exceed the signal of the target compound. The identification is performed in three steps. In the first step, the signatures contained in the matrix are fitted to the brightness temperature spectrum. In the next step the contributions of all fitted signatures (i.e. interferents, ozone, water, and baseline) except the signature of the target compound are subtracted from the measured spectrum. To decide if the target compound is present, the coefficient of correlation between the corrected spectrum, i.e. the result of the subtraction, and a reference spectrum is calculated in a compound specific number of spectral windows. Moreover, the coefficient of correlation between the fitted spectrum and the measured spectrum is calculated. The signal to noise ratio is calculated by division of the maximum brightness temperature difference caused by the target compound by the noise equivalent temperature difference of the spectrum. If all coefficients of correlation and the signal to noise ratio are greater than compound specific threshold values the target compound is identified. This calculation is performed for three different column densities of the target compound. Optionally, the difference between a background spectrum and the spectrum of the cloud can be analyzed in addition to the direct analysis of brightness temperature spectra. If the scanning system is used, the difference between spectra of adjacent fields of view may be analyzed as proposed in reference 14. A detailed description of the algorithm and a review of other methods are given in reference 7.
2.3 Quantification

2.3.1 Introduction

If the cloud temperature is equal to the temperature of layer 1 or if the transmittance of layer 1 is high in the spectral range that is used for the analysis ($\tau_1 \approx 1$), Eq. (4) or Eq. (1) respectively can be rearranged to give

$$\tau_2 = \frac{L_1 - B_2}{L_b - B_2}. \quad (7)$$

There are several methods to determine the quantities $L_b$ (the background radiance) and $B_2$ (i.e. the temperature of the cloud) in Eq. (7). Under the assumption of thermal equilibrium, $L_b$ and $B_2$ may be determined by measurement of a background spectrum and by measurement of the spectrum of a black plate at ambient temperature, respectively.\(^{15,16}\) It is also possible to determine these quantities by analysis of the measured spectrum. Under the assumption of thermal equilibrium, the cloud temperature may be estimated by calculation of the mean of the brightness temperature in a spectral range within which the atmosphere is opaque\(^{17}\). Another method is the analysis of transitions of CO\(_2\) or H\(_2\)O by nonlinear fitting. The radiance of the background may be determined by calculation of the baseline of the brightness temperature spectrum in the spectral range of the transitions of the molecules of the cloud\(^7\). The advantage of this method is, that no background spectrum is required for the analysis. This is the standard method implemented in the data analysis program of the scanning system for the determination of the brightness temperature of the background, the ambient air temperature and for quantification for high transmittance $\tau_2$.

The column density $c_1$ may be calculated by various methods applying the Lambert-Beer law:

$$r(\sigma) = \exp(-\sigma(\sigma)c_1), \quad (8)$$

$$A(\sigma) = -\log_{10}(r(\sigma)) = \ln(10)^{-1}\sigma(\sigma)c_1, \quad (9)$$

where $\sigma(\sigma)$ is the absorption coefficient of the compound and $c_1$ is the column density. Eq. (9) is the definition of absorbance $A(\sigma)$. The error in the column density retrieved by the methods described above caused by erroneous estimation of the cloud temperature increases when the transmittance and the radiance difference decrease. The strong increase with decreasing transmittance is caused by the exponential dependence of the transmittance on the column density. An estimation of this error is presented in reference 15.

Moreover, an additional systematic error caused by the finite resolution of the measured spectrum is introduced if Eq. (9) is applied using the measured radiance. Eq. (9) is valid for monochromatic transmittance. The effect of finite spectral resolution on the quantification by Eq. (9), an apparent deviation from the Lambert-Beer law, has been described in various publications (see for example reference 18). This error is increasing with decreasing transmittance and decreasing spectral resolution. For passive remote sensing of pollutant clouds the lowest resolution that fulfills the requirements of selectivity of the specific application is used in order to obtain the best possible signal to noise ratio. Thus, the width of the instrumental line shape $ILS(\sigma)$ is often greater than the width of the transitions that are analyzed and the effect of finite spectral resolution must be considered in order to calculate accurate column densities.

2.3.2 Quantification by Nonlinear Modeling

The quantification is performed by minimization of the difference between the measured spectrum $L_{\text{meas}}$ and a spectrum calculated using a model, $L_{\text{model}}$:

$$\sum_{i=1}^{N} \left( l_{i,\text{model}} - l_{i,\text{meas}} \right)^2 = \min \quad (10)$$

$N$ is the number of spectral elements. In order to model the measured spectrum, a radiative transfer model for the radiance at the entrance aperture of the interferometer and a model describing instrumental effects are required.
For an ideal interferometer, operating with perfectly collimated radiation, the instrumental line shape ILS is given by the Fourier transform of the apodization function, \( A_\lambda \). In practice, especially in the case of interferometers optimized for high throughput, non-parallel rays from a finite source, which subtends a solid angle \( \Omega \), reach the detector, causing a broader ILS and a frequency shift\(^{19} \). The spectral radiance \( L_{\text{meas}}(\sigma) \) in a small range around a wavenumber \( \sigma_i \), measured by an FTIR spectrometer is a linear function of the convolution

\[
S(\sigma) \ast ILS^i(\sigma).
\] (11)

Here \( S(\sigma) \) is the true spectrum, i.e. the spectrum measured with infinite optical path difference and collimated radiation. \( ILS^i \) is the instrumental line shape at wavenumber \( \sigma_i \), \( \ast \) is the convolution operator. The instrumental line shape of a real FTIR spectrometer with a finite limiting aperture may be approximated by

\[
ILS^i(\sigma) = A^i_\lambda(\sigma) \ast \delta(\sigma - \Delta \sigma_i),
\] (12)

where \( A^i_\lambda(\sigma) \) is a function describing the effect of broadening, and \( \delta(\sigma - \Delta \sigma_i) \) is a delta (Dirac) function, describing the frequency shift. In the case of a homogeneous source and an ideal interferometer (ideal mirrors etc.), \( A^i_\lambda(\sigma) \) is an even boxcar function of total width \( \frac{\sigma_\lambda \Omega}{2\pi} \), and the frequency shift is\(^{19} \) \( \Delta \sigma_i = -\frac{\sigma_i \Omega}{4\pi} \).

Equations (1) and (2) may be used to model radiative transfer but they contain a large number of parameters and unknown optical properties like the biconical reflectance function of the surface and the temperatures and the composition of higher layers of the atmosphere. These quantities are not the target of the retrieval and the calculation of the radiance from the sky is a time consuming process. Because the goal of the development of the quantification method developed in this work is a fast algorithm, Eq. (4) with one additional layer between the cloud and the background is used. For the most important case, the case of thermal equilibrium, Eq. (1) and Eq. (4) are equivalent. However, if the background of the measurement is the sky, emission signatures of higher, colder layers of the atmosphere dominate the background spectrum. Thus, it is advantageous to use Eq. (4). The additional layer models the signature of ozone, which is the most important atmospheric interferent in the atmospheric window (800 - 1200 cm\(^{-1} \)), besides water. The radiance of the background is modeled by the spectrum of a blackbody in combination with a superposition of broad Gauss functions centered at different frequencies. The emittance of many background materials is a slowly varying function of the frequency. Thus, broad Gauss functions are suitable to model deviations from the ideal blackbody spectrum.

The transmittance of the cloud is calculated by convolution of a spectrum of monochromatic transmittance with the normalized ILS.

\[
\tau^c_{\lambda}(\sigma) = \tau(\sigma) \ast ILS^i(\sigma)
\] (13)

The monochromatic transmittance of the cloud \( \tau(\sigma) \) is calculated with the use of Eq. (8). If the target species is contained in the HITRAN\(^{20} \) Database, absorption coefficients are calculated with the use of FASCODE\(^{21} \) under conditions of the standard atmosphere\(^{22} \). Alternatively a measured high-resolution spectrum may be used. Because the maximum range for the analysis is given by the atmospheric window, the ILS of the spectrometer is approximated by a constant value of \( \sigma_i \) (\( \sigma_i = 1000 \text{ cm}^{-1} \)). In order to minimize computation time, the calculation of all other layers is performed with the spectral resolution of the measurement. If the transmittance of the other layers is high, the transmittance calculated using Eq.(8) with low-resolution absorbance spectra is a good approximation. The (apparent) absorption coefficients are calculated by a convolution of spectra of monochromatic absorption coefficients with the ILS. The transmittance of layer 1 is calculated using a spectrum of the standard atmosphere calculated by FASCODE/HITRAN and a convolution with the ILS. In this work, a spectral resolution of 4 cm\(^{-1} \) is used. Eq. (10) may be written as
\[
\sum_{i=1}^{N} \left( L_{i, \text{model}}(\sigma_i, T_1, T_M, l_1, c_{l2}, c_{lM}, T_{brbg}, b_1, b_p) - L_{i, \text{meas}} \right)^2 = \min ,
\]

where \( M \) is the number of atmospheric layers, \( T_j \) is the temperature of layer \( j \), \( l_1 \) is the effective length of layer 1, \( c_{l_j} \) is the column density of layer \( j \), \( T_{brbg} \) is the mean of the brightness temperature of the background, and \( b_1, b_p \) are the coefficients of \( P \) Gauss functions. The starting values are determined by the linear model, which is used for identification (see 2.2). If the compound is identified and the signal, i.e. the maximum brightness temperature difference caused by the compound is greater than a threshold value (currently 1 K), the best-fit parameters are determined by the Levenberg-Marquardt method. Moreover, methods for the calculation of the best-fit parameters of the ILS have been developed. For the approximation of \( \Omega \), Eq. (12) becomes

\[
\sum_{i=1}^{N} \left( L_{i, \text{model}}(\sigma_i, T_1, T_M, l_1, c_{l2}, c_{lM}, T_{brbg}, b_1, b_p, \Omega) - L_{i, \text{meas}} \right)^2 = \min .
\]

To determine the frequency shift,

\[
\sum_{i=1}^{N} \left( L_{i, \text{model}}(\sigma_i, T_1, T_M, l_1, c_{l2}, c_{lM}, T_{brbg}, b_1, b_p) - L_{i, \text{meas}}(f, x_{oi}) \right)^2 = \min
\]

is used. Here \( x_{oi} \) is the wavenumber of spectral element \( i \) as determined by the previous frequency calibration (the starting value for wavenumber \( x_i \)), \( f \) is the correction factor and \( x_i = f \times x_{oi} \). Because both methods require more calculation time than the calculation with a constant ILS, these methods are only applied if necessary, for example if a modification of the interferometer is carried out or a different interferometer is used.

The relation that allows quantification by nonlinear modeling of a small spectral range at ambient temperatures and small temperature differences is the exponential dependence of the transmittance on the column density. The ratios of signatures in the transmittance spectrum (i.e. \( 1 - t \)) of transitions with different absorption coefficients are invertible functions of column density. However, if the transmittance of the cloud is high \((t_2 \approx 1)\) in the whole spectral range, the ratios of signatures with different absorption coefficients in the transmission spectrum are almost constant. Thus, the column density calculated by the method described in the previous section is used, if the resulting transmittance retrieved by the nonlinear method is above a compound specific threshold value \( \tau_{2min} \). In the case of ammonia, this value is \( \tau_{2min} = 0.85 \).

### 3. THE SCANNING INFRARED REMOTE SENSING SYSTEM

The scanning infrared remote sensing system is comprised of an interferometer (OPAG 22, Bruker Saxonia Analytik GmbH, Leipzig, Germany), an azimuth-elevation-scanning mirror actuated by stepper motors, a data processing and control system with a digital signal processor (FTIR DSP), an image processing system (Video DSP), and a personal computer (FIG. 2).

For the visualization of pollutant clouds, the scanning mirror is sequentially set to all positions within the field of regard. The size and the direction of the field of regard and the spatial resolution (i.e. the angle between adjacent fields of view) are variable. Each interferogram measured by the interferometer is recorded by the FTIR DSP system. The Fourier transformation is performed by the DSP and the spectrum is transferred to the PC. The spectrum is analyzed using the identification method described previously. The results are visualized by a video image, overlaid by false color images. For each target compound of the spectral library, images of the coefficient of correlation, the signal to noise ratio, the brightness temperature of the background, the difference between the temperature of the ambient air and the brightness temperature of the background, and the noise equivalent column density are produced (FIG. 3). Simultaneously with the analysis and visualization of one interferogram by the FTIR DSP and the PC, the scanning mirror is set to the next position and the next interferogram is recorded. After the measurement of the field of regard is completed, the column densities of all directions in which a compound has been identified may be calculated and an additional false color image is displayed.
The video processing system records and analyzes images of a video or an infrared camera. Different modes of operation are available. In the standard mode, the video image is used for visualization as described above. Moreover, the image of an infrared camera can be analyzed in order to align the scanner automatically to backgrounds that yield a low limit of detection (i.e. backgrounds that yield a large temperature difference).

### Table 1: Specifications of the scanning FTIR system.

<table>
<thead>
<tr>
<th>Specification</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Spectral range</td>
<td>680 – 1500 cm⁻¹</td>
</tr>
<tr>
<td>Maximum spectral resolution</td>
<td>0.5 cm⁻¹</td>
</tr>
<tr>
<td>Resolution for remote sensing</td>
<td>4 cm⁻¹</td>
</tr>
<tr>
<td>Étendue</td>
<td>0.008 sr cm²</td>
</tr>
<tr>
<td>Field of view</td>
<td>30 mrad</td>
</tr>
<tr>
<td>Field of view with telescope</td>
<td>7.5 mrad</td>
</tr>
<tr>
<td>$\text{NESR} (\Delta \sigma = 4 \text{~cm}^{-1}, \text{<del>0.1</del>s})$</td>
<td>$3.3 \times 10^{-7}$ W/cm² sr cm⁻¹</td>
</tr>
<tr>
<td>Scan rate ($\Delta \sigma = 4 \text{~cm}^{-1}$)</td>
<td>6 spectra/s</td>
</tr>
<tr>
<td>Field of regard</td>
<td>285° x 80°</td>
</tr>
<tr>
<td>Maximum angular resolution of the scanner</td>
<td>0.1°</td>
</tr>
<tr>
<td>Power consumption</td>
<td>60 W (without PC)</td>
</tr>
</tbody>
</table>

4. **EXPERIMENTAL**

In order to verify the quantification method, a gas cell was mounted in front of the entrance window of the interferometer. The length of the stainless steel cell was 0.15 m, the material of the windows was NaCl. All measurements were performed with a spectral resolution of 4 cm⁻¹ and triangular apodization. The experiments were performed with ammonia, because ammonia is one of the most important target compounds of the system. Different sample concentrations were prepared by mixing the appropriate volumes of ammonia and air in a sample bag. A heated black plate (active area 25 x 25 cm²) was used for the radiometric calibration. Moreover, the black plate was used as radiation source behind the gas cell in laboratory measurements. For measurements with an atmospheric optical path, the scanning mirror is used to align the field of view to different backgrounds. This setup is shown in FIG. 4.
results of field measurements are presented. Ammonia that evaporated from a meadow, which was treated with liquid manure was measured by the scanning FTIR system. Column densities are expressed as equivalent concentration path length products in ppm m (288.15 K, 101325 Pa).

FIG. 4. Experimental setup.

FIG. 5. Measured spectrum after subtraction of the spectrum of a blackbody at the brightness temperature of the background (solid), result of the fitting calculation (dashed). Lower curve: difference (solid, offset subtracted).
5. RESULTS AND DISCUSSION

FIG. 5 shows a spectrum of 750 ppm m ammonia in air in front of the black plate (1 scan, $T_{bb} = 305$ K). Moreover, the spectrum calculated with the best-fit parameters and the difference are displayed. The calculation was performed in the range 775 – 1250 cm$^{-1}$. Because the radiance spectrum does not have a constant baseline over this spectral range, the spectrum of a blackbody at the brightness temperature of the background was subtracted. The retrieved brightness temperature of the background was $T_{bbg} = (304.081 \pm 0.076)$ K, the difference between the temperature of the sample and the brightness temperature of the background $\Delta T$ was $(-3.46 \pm 0.13)$ K, the retrieved $cl$ of the spectrum shown in FIG. 5 was $(764 \pm 43)$ ppm m. Standard deviations were calculated for a single measurement (1 scan) using 100 measurements.

![Graph showing retrieved $cl$ as a function of the true $cl$. Error bars represent standard deviations. Dashed line: $cl_{NH3} = cl_{retrieved}$.](image)

In order to verify the linearity, measurements with different column densities were performed. FIG. 6 shows the retrieved column density as a function of the true column density (100 measurements, 1 scan, $T_{bb} = 305$ K). The calculation was performed in the range 775 – 1250 cm$^{-1}$. The sample temperature was approximately room temperature. Thus, the retrieved difference between the brightness temperature of the background and the temperature of the sample varied in the course of the experiments. The retrieved differences were $\Delta T_{500ppmm} = (-2.97 \pm 0.35)$ K, $\Delta T_{450ppmm} = (-2.52 \pm 0.20)$ K, $\Delta T_{750ppmm} = (-3.52 \pm 0.13)$ K, $\Delta T_{1500ppmm} = (-3.91 \pm 0.07)$ K, and $\Delta T_{3000ppmm} = (-3.06 \pm 0.04)$ K. The equation of the regression line was $cl_{ret} = 0.994 \times cl_{true} + 30$ ppm m. The coefficient of correlation was 0.9997.

The graph on the left side of FIG. 7 shows the results of 500 measurements of a sample of ammonia with different temperatures of the black plate. The retrieved column density is plotted versus the difference between the retrieved temperature of the sample and the brightness temperature of the background. As the temperature difference increases, the variance of the retrieved column density decreases and the retrieved values converge to the mean value. The retrieved column density of the last 400 measurements ($|\Delta T| > 6.5$ K) was $(721 \pm 13)$ ppm m. The calculation was performed in the range 775 – 1250 cm$^{-1}$. Moreover, FIG. 7 shows the results of 600 measurements of the same sample with atmospheric optical paths. The scanner system was used to scan an area with different backgrounds. The measurements include measurements with the sky as background. The retrieved column density was $(703 \pm 36)$ ppm m. The calculation was performed in the range 900 - 1000 cm$^{-1}$. The small spectral range minimizes the effect of atmospheric interferents on the quantification. On the other hand the variance of the retrieved column densities increases because less data points are used for the calculation.
FIG. 7. Left: Quantification of a sample of ammonia in air at different temperatures of the black plate. Right: Results of the quantification algorithm for 600 spectra of the same sample with different atmospheric optical paths and different backgrounds including the sky. The retrieved column density is plotted versus the difference between the retrieved temperature of the sample and the brightness temperature of the background.

FIG. 8. Results of measurements of ammonia, which evaporated from the meadow. The coefficient of correlation is only displayed if ammonia is identified. The area that was scanned is highlighted by the rectangle.
FIG. 8 and FIG. 9 show results of measurements of ammonia that evaporated from a meadow, which was treated with liquid manure. The image of the column density retrieved by the nonlinear fitting algorithm is displayed in FIG. 9. Four interferograms were added before the Fourier transformation. The mean of the column density was $c_I = 147 \text{ ppm m}$, the standard deviation was 31 ppm m. The retrieved temperature of ammonia was $T_2 = (282.2 \pm 1.4) \text{ K}$. This result is in good agreement with the ambient air temperature retrieved by calculation of the mean of the brightness temperature spectrum in the range 680 – 690 cm$^{-1}$, $T_1 = (282.01 \pm 0.84) \text{ K}$ and the temperature of the ambient air measured with a conventional thermometer, $T_{am} = 282.6 \text{ K}$. The concentration of ammonia was $(700 \pm 150) \text{ ppb}$.

6. CONCLUSION

The scanning remote sensing system allows identification, visualization, and quantification of pollutant clouds. The quantification algorithm allows quantification without assumptions about the cloud temperature. Because the method models the influence of the instrument on the signal, the retrieved column density is proportional to the true column density over a wide range. The apparent deviation from the Lambert-Beer law at low spectral resolution, which arises from the finite width of the instrumental line shape, is not observed. If the instrumental line shape is well characterized, and the absorption coefficients of the target compound are known, the only calibration procedure required is the radiometric calibration of the spectrometer.

Recently, the system has been extended with a positioning system (GPS) and an electronic compass. The position of the system, the field of regard, and the directions in which a target compound was identified are automatically marked in a map. If a cloud is measured from different positions, the contaminated area is calculated and visualized in the map. A possible future application of the quantification method presented in this work is the calculation of concentration maps using a tomographic reconstruction algorithm.

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