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Master of Science

Development of a Tomographic Atmospheric Monitoring System based on Differential Optical Absorption Spectroscopy

Thesis plan submitted in partial fulfillment of the requirements for the degree of

Doctor of Philosophy in **Biomedical Engineering**

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Contents

1	Inti	roduction	1
	1.1	Background and Motivation	1
		1.1.1 Background	1
		1.1.2 The Problem	1
		1.1.3 Objectives	2
		1.1.4 Methods	2
2	Res	earch Question	5
	2.1	Problem Introduction	5
	2.2	Research Question	6
	2.3	Hypothesis and Approach	7
3	Lite	erature Review	9
	3.1	Air pollution and pollutants	9
	3.2	Tomographic algorithms and reconstruction techniques	10
		3.2.1 Introduction	10
		3.2.2 Initial Considerations	11
		3.2.3 The Fourier Slice Theorem	12
		3.2.4 The Filtered BackProjection Algorithm	13
	3.3	DOAS	15
	3.4	DOAS tomography	15
		3.4.1 Active DOAS tomography	15
		3.4.2 Passive DOAS tomography	15
4	Res	earch Methodology	17
	4.1	Aimed contribution	17
	4.2	Detailed work plan and scheduling	17
	4.3	Validation methodology	17
	4.4	Integration with other research activities	17

Bibliography 19

List of Figures

3.1	A schematic representation of a projection	11
3.2	Schematic representation for coordinate setting	12
3.3	The FST, a schematic representation.	14
3.4	Schematic representation of an equiangular fan beam projection	15

List of Tables

2.1	Main research question	7
2.2	Secondary research questions	7

Glossary

aliquam tincidunt urna. Nulla ullamcorper vestibulum turpis. Pellentesque

cursus luctus mauris.

computer An electronic device which is capable of receiving information (data)

in a particular form and of performing a sequence of operations in accordance with a predetermined but variable set of procedural instructions (program) to produce a result in the form of information

or signals.

cras viverra metus rhoncus sem. Nulla et lectus vestibulum urna fringilla ultrices.

Phasellus eu tellus sit amet tortor gravida placerat.

donec nonummy pellentesque ante. Phasellus adipiscing semper elit. Proin fermen-

tum massa ac quam. Sed diam turpis, molestie vitae, placerat a,

molestie nec, leo.

integer sapien est, iaculis in, pretium quis, viverra ac, nunc. Praesent eget sem vel

leo ultrices bibendum. Aenean faucibus.

lorem ipsum dolor sit amet, consectetuer adipiscing elit. Ut purus elit, vestibulum

ut, placerat ac, adipiscing vitae, felis. Curabitur dictum gravida

mauris.

maecenas lacinia nam ipsum ligula, eleifend at, accumsan nec, suscipit a, ipsum. Morbi

blandit ligula feugiat magna. Nunc eleifend consequat lorem.

morbi ac orci et nisl hendrerit mollis. Suspendisse ut massa. Cras nec ante.

Pellentesque a nulla. Cum sociis natoque penatibus et magnis dis

parturient montes, nascetur ridiculus mus.

morbi dolor nulla, malesuada eu, pulvinar at, mollis ac, nulla. Curabitur auctor

semper nulla. Donec varius orci eget risus. Duis nibh mi, congue eu, accumsan eleifend, sagittis quis, diam. Duis eget orci sit amet orci

dignissim rutrum.

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tortor sed accumsan bibendum, erat ligula aliquet magna, vitae

ornare odio metus a mi.

nam dui ligula, fringilla a, euismod sodales, sollicitudin vel, wisi. Morbi auc-

tor lorem non justo.

name arcu libero, nonummy eget, consectetuer id, vulputate a, magna. Donec ve-

hicula augue eu neque. Pellentesque habitant morbi tristique senectus et netus et malesuada fames ac turpis egestas. Mauris ut leo.

nulla malesuada porttitor diam. Donec felis erat, congue non, volutpat at, tincidunt

tristique, libero. Vivamus viverra fermentum felis.

sed lacinia nulla vitae enim. Pellentesque tincidunt purus vel magna. Integer

non enim. Praesent euismod nunc eu purus. Donec bibendum quam

in tellus.

Acronyms

AP Air Pollution

CT Computed Tomography

DOAS

EPA Environmental Protection Agency (United States)

Differential Optical Absorption Spectroscopy

FBP Filtered BackProjection
FFF Forest Fire Finder

FST Fourier Slice Theorem FT Fourier Transform

11 Tourier Transform

ICE Internal Combustion Engine IFT Inverse Fourier Transform

ML Machine Learning

PM Particulate Matter

ROI Region Of Interest RQ Research Question

SLR Systematic Literature Review

VOC Volatile Organic Compound

WHO World Health Organization

Symbols

*

Introduction

1.1 Background and Motivation

1.1.1 Background

The idea behind this thesis was born in 2015, at NGNS-IS (a Portuguese tech startup). At the time, the company's flagship product was the Forest Fire Finder (FFF), which was a forest fire detection system that performed a spectroscopical analysis of the atmosphere and then, through some Machine Learning (ML) techniques, could detect the presence of a smoke column above the horizon and alert the operators.

The growing importance of Air Pollution (AP) in today's society, and the fact that the system was already scanning the atmosphere for some chemical components originated and motivated the idea behind this thesis. Although FFF was already a spectroscopic system, it was constructed to operate in remote and inhospitable locations, and its design had had no spatial constraints into account. In addition, the system scanning method meant that it was not appropriate for pollution measurement, as it could only detect a mean pollutant column density for each spectrum it took. A truly useful monitoring tool would be able to map these pollutants concentration, thus retrieving the same kind of information as a network of in-situ electro-chemical sensors.

There were, at the time, a few research projects that aimed to bring this kind of capacity to Differential Optical Absorption Spectroscopy (DOAS) based systems. However, there was ample room for improvement, as it was made clear by the lack of commercial systems and the scarcity of the literature on the subject. Realizing the type of research that accomplishing this project would entail, the company decided to publish this PhD Project, in a tripartite consortium with FCT-NOVA and the Portuguese Foundation for Science and Technology.

1.1.2 The Problem

Air Pollution (AP) is one of the grave concerns of modern day western society, with many decades worth of research proving that it can have a pronounced negative effect on human, animal and plant life, as shown in Section ??. On humans, it has been shown to significantly increase risk of cardiovascular, pulmonary and even neuropsychiatric diseases [2, 4, 10]. Its implications on ecosystems are remarkably complex and difficult to quantify, but nonetheless extremely important, and have a huge impact on biodiversity [11].

Knowing all this brings us the responsibility of at least trying to mitigate some of these adverse consequences of the spectacular progress that we have achieved in the few last centuries. But we cannot act unless we also know what we must do; and to know this, we must have the devices and means to employ them that allow us to measure AP, to trace it and to understand it, so that our actions against it are effective.

1.1.3 Objectives

The overarching goal of this thesis was to theorize and design a bidimensional mapping tool for trace atmospheric pollutants such as NO_x and SO_x , using DOAS as the measurement technique. In order to maximize commercial value (and viability), the system had to be small and mobile. During the research, several "micro-objectives" appeared regularly. Some were kept and incorporated in the workplan, others discarded after initial exploration. The main secondary objectives were:

- To use a tomographic approach for the mapping procedure;
- To ensure the designed system would be small and highly mobile;
- To use a single collection point, minimizing material costs for the technology.

These objectives allowed setting several research questions, which are introduced in Section 2.2.

1.1.4 Methods

To address these goals, I assumed the development of this thesis to be essentially split in two parts, which are to be explored simultaneously. They can (coarsely) be addressed as *tomography* and *instrumentation*. On the tomography side, it will be necessary to study what are the more appropriate algorithms (and what type of tomography), how they can be physically deployed (i.e., the problem's geometry) and what type of reconstruction method is the more favorable. Regarding the instrumentation, there are also several points that need considering: decisions are required with regard to the mechanics, the controls and the optical components of the final system. A more detailed discussion of these topics can be found in Section 2.3 and Chapter ??.

One of the most important steps in the development of this work is designing and implementing a simulation software platform, that allows the validation of the acquisition strategy, the geometry selection and the reconstruction approach. This endeavor will also be of critical importance for component selection, since it will define the component requirements for the whole system. On the optical instrumentation side, this project will require optimizing FFF's optical assembly. Although similar in purpose and types of components, this assembly is significantly larger than what is acceptable for this project and needs redesigning.

Research Question

2.1 Problem Introduction

AP is a very important topic of discussion in the current days, with scientists and researchers around the globe being very well aware of the potential effects it can have on the health of individuals and populations across all ecosystems. After climate change (one of the largest capital threats to life on Earth, perhaps just behind nuclear apocalypse), AP is the biggest environmental concern for Europeans, and Europe's single largest environmental health hazard [**EEA2016**]. It has also been established by many authors as a major cause of premature death, cardiopulmonary disease onset and hospital visits [**EEA2007**, **EEA2016**, 4, 13].

Even in spite of all the trouble it causes, defining AP can be a challenge. In fact, its effects and presence is so all-encompassing, that it would be fair to say that its definition changes with the angle with which one looks upon it. The United States Environmental Protection Agency (EPA) defined Air Pollution (AP) as "the presence of contaminants or pollutant substances in the air that interfere with human health or welfare, or produce other harmful environmental effects" [12]. This is (perhaps intentionally) a very broad definition, too broad to avoid vagueness. It does introduce a key concept: the term pollutant, which needs be discussed in order to complete the definition above.

It would be very hard to find someone who did not have an almost instinctive idea of what a pollutant is. We know something is amiss when we notice our air is full of smoke or smells strange, but our senses are not enough. There are many chemical components that are untraceable by unaided humans, and some that are only detected by our noses and eyes at concentration levels which are above the threshold where they can damage our health. This makes the task of separating pollutants from non-pollutants a non-trivial one. If we cannot rely solely on our senses to detect them, then it is up to the scientists and engineers to create methods that allow us to do so. Whats more, we must also rely on them to understand how can a normally harmless substance be a pollutant, depending on the circumstance. For instance, nitrous compounds are

traditionally beneficial to the soils and cultures, but they can and do cause pulmonary and cardiovascular complications in humans [2, 4, 10].

Context matters to pollutants. The toxic nature of a certain chemical only is revealed when someone or something gets exposed to it. Even then, there are exposure levels which do not bear any effects, good or bad. At these levels, a pollutant is but an impurity. There are too many potential pollutants in our modern day world to list here, but several reports make special mention to six pollutions, which are identified as being the major contributors to Air Pollution complications.

- Particle Matter (PM);
- Ground level ozone (O₃);
- Carbon monoxide (CO);
- Sulfur Oxides (SO_x);
- Nitrous Oxides (NO_x);
- Lead (Pb).

Exposure to these pollutants has different effects on humans, ranging in seriousness from skin irritation to neuropsychiatric complications, depending on dose and on the time the exposure lasts.

2.2 Research Question

In Chapter 1, I have introduced the reasons which led NGNS-IS to pursue the development of an atmospheric monitoring system, and that what set it apart from other systems was the ability to spectroscopically map pollutants concentrations using tomographic methods, thus defining a primary objective for this thesis.

Two secondary objectives were born from the necessary initial research, which had a very heavy influence over the adopted methods:

- To use a tomographic approach for the mapping procedure;
- To ensure the designed system would be small and highly mobile;
- To use a single light collection point, minimizing material costs.

Taking all the above into account, we arrive at the main Research Question (RQ), presented in Table 2.1.

This is the main research question. It gave rise to four other more detailed research questions. These secondary questions allow a better delimitation of the work at hand and are important complements to RQ1. This questions are presented in Table 2.2

Table 2.1: Main research question.

RQ1 How to design a miniaturized tomographic atmosphere monitoring system based on DOAS?

Table 2.2: Secondary research questions.

RQ1.1 What would be the best strategy for the system to cover a small geographic region?

RQ1.2 What would be the necessary components for such a system?

RQ1.3 How will the system acquire the data?

RQ1.4 What should the tomographic reconstruction look like and how to perform it?

2.3 Hypothesis and Approach

This work is based on the hypothesis that a system such as the one described in Chapter 1, which responds to the RQ in Table 2.1 and Table 2.2 can be achieved by careful selection of mathematical tomographic algorithms and instrumentation that is able to implement them correctly.

The first step in answering the entirety of the research questions should be to answer RQ1.1. In fact, it is not possible to make any other decision before this matter is settled. As with any technical problem, there are several ways to create a tomographic atmospheric monitoring tool. However, each and every one of them implies some kind of compromise, which determines the system's capabilities and requirements. Will the system use retro-reflection? Shall it move during the measurement? These are the kind of questions that determine the whole project.

When the measurement strategy is determined, one could start picking parts and components. However, a better first approach would be designing a software simulation. This simulator must include all major system features, so that it correctly mimics reality and is therefore able to mathematically validate the acquisition and reconstruction approach. The results obtained from the simulation will then dictate mechanical and control requirements.

One other aspect that needs addressing is the optical section. As mentioned before, the system will be inspired in FFF's basic optical capabilities. However, the smoke detector was not conceived with spatial restrictions in mind. This important set of components will thus need redesigning, so that it is in line with the size objectives of the new system.

Literature Review

In this chapter, I provide a literary review on the three most important subjects for the work of this thesis: AP, tomographic algorithms and DOAS tomography instrumentation.

3.1 Air pollution and pollutants

As stated in Section 2.2, the definition of AP is dependent on the context. Here, I will focus especially on the effects of pollutants on human health. Whether these effects are the most significant problems stemming from AP is debatable (climate change is mostly caused by anthropogenic production of greenhouse gases, which are pollutants) but for this system and its intended uses, health effects are definitely more prominent. Human health implications of a polluted atmosphere are documented in very numerous studies throughout the literature. In this document, I will only present a small number of representative reviews and reports.

In 2004, WHO published a report summarizing what was then the most recent information on health effects of air pollution over Europe. This review concluded that, even with all the regulations on AP put in place by the European authorities, its levels were still posed a considerable burden on health throughout Europe [13].

Although there are several hundred potentially harmful components already that have already been found in the atmosphere, this report addresses only PM, ground level Ozone and Nitrogen Dioxide. As many other studies had found, this Systematic Literature Review (SLR) identified several short-term and long-term exposure effects for the three pollutants. The study found that short-term exposure to all three substances were responsible for an increase in mortality and hospital admissions, and that both PM and O_3 increased the population's usage of medication. Long-term exposure to all three components have adverse pulmonary effects, but PM have many other negative effects. The most important of them a reduction in life expectancy, which the authors attribute to cardiopulmonary mortality and lung cancer.

Particulate Matter are described as airborne solid particles or droplets. These

particles vary in size, origin and composition, however, it is usual to classify them by size, since that is what governs particle deposition in the respiratory system. Urban PM are usually divided into three categories: coarse, fine and ultrafine. Convention dictates that coarse particles have an aerodynamic diameter of less that $10\mu m$ (PM $_{10}$), fine particles less than $2.5\mu m$ (PM $_{2.5}$). In this review, the authors stated that the role that ultrafine particles play in human health is still undetermined, but they noted that coarse and (especially) fine particles are highly correlated with an increase in mortality and the prevalence of respiratory syndromes [13].

Ground-level ozone is produced as a result of a chemical reaction between nitrous oxides and Volatile Organic Compounds (VOC), which can be emitted either by natural source or by human-related activities. O_3 is a powerful oxidant, and can easily accept electrons from other molecules. In the respiratory tract, this chemical destroys double bonds of the fatty acids in the surface of its lining. The process leads to the deposition of several substances like aldehydes and hydrogen peroxide, resulting in impaired cell function. Ozone is toxic at concentrations that occur in urban areas around the world. On a more ecological level, it has been established that O_3 is responsible for a decrease in the trees' capabilities to assimilate carbon, which can result in deforestation.

Nitrous Oxides originate from the use of Internal Combustion Engines (ICE) for energy production and especially movement. It is usually used as an indicator for the presence of heavy traffic. The compound acts in a more subtle way than Ozone, described above. It increases the risk of respiratory infections and can cause pulmonary edema. Moreover, it has an effect on the immunological system, impairing the ability of T-lymphocytes to address microbiological threats like viruses. Nitrous oxides are also known to affect weaker populations, like children and the elderly.

3.2 Tomographic algorithms and reconstruction techniques

3.2.1 Introduction

Tomography is the cross-sectional imaging of an object through the use of transmitted or reflected waves, captured by the object exposure to the waves from a set of known angles. It has many different applications in science, industry, and most prominently, medicine. Since the invention of the Computed Tomography (CT) machine in 1972, by Hounsfielf [5], tomographic imaging techniques have had a revolutionary impact, allowing doctors to see inside their patients, without having to subject them to more invasive procedures [9].

Mathematical basis for tomography were set by Johannes Radon in 1917. At the time, he postulated that it is possible to represent a function written in $\mathbb R$ in the space of straight lines, $\mathbb L$ through the function's line integrals. A line integral is an integral in which the function that is being integrated is evaluated along a curved path, a line.

ations

In the tomographic case, these line integrals represent a measurement on a ray that traverses the Region Of Interest (ROI). Each set of line integrals, characterized by an incidence angle, is called a projection (see Figure 3.1). To perform a tomographic reconstruction, the machine must take many projections around the object. To the set of projections arranged in matrix form by detector and projection angle, we call sinogram. All reconstruction methods, analytical and iterative, revolve around going from reality to sinogram to image [1, 3, 6–9].

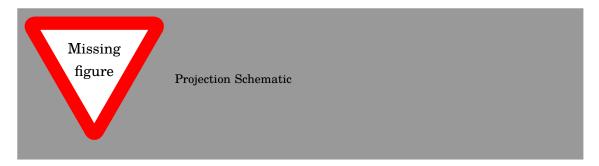


Figure 3.1: A schematic representation of a projection.

There are two broad algorithm families when it comes to tomographic reconstruction, regarding the physics of the problem. The problem can involve either non-diffracting sources (light travels in straight lines), such as the X-Rays in a conventional CT exam; or diffracting sources, such as micro-waves or ultrasound in more research-oriented applications. In this document, I will not address the latter family, since I will not be applying them in my work. In the next few paragraphs, I will discuss the first family of algorithms, and describe how an image can be reconstructed from an object's projections when the radiation source is non-diffracting.

Let's consider the case in which we deal with a single ray of solar light entering the atmosphere at a given point. Since the atmosphere contains numerous absorbents and comparable atmospheric effects, the ray changes from the point where it enters the atmosphere to the point at which it is measured by a detector. Total absorption will depend on the pollutant species, their cross-section and their concentration, since it obeys Lambert-Beer's law. Looking from another angle, this absorption is also the line integral that we will use to reconstruct our image. With DOAS, it is possible to measure several pollutants at the same time, but for simplicity (and since it is one of the most studied compounds in the field), let's consider that the single pollutant in our atmospheric mixture is NO_2 .

3.2.2 Initial Considerations

The problem of tomographic reconstruction can be approached in a number of ways, depending mostly on the authors. In my literary search, I have found that Kak and Slaney [9] have certainly explained this problem in one of the clearer ways available.

Therefore, I shall base the rest of my presentation in their writings, and complement with other authors' notes wherever necessary.

Considering the coordinate system displayed in Figure 3.2. In this schematic, the object is represented by the function f(x, y). The (θ, t) parameters can be used to define any line in this schematic. Line AB in particular can be written:

$$x \cdot \cos(\theta) + y \cdot \sin(\theta) = t \tag{3.1}$$

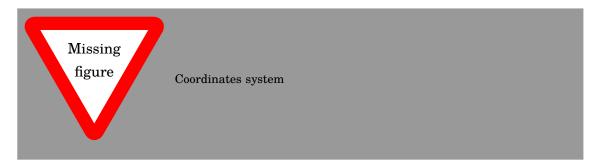


Figure 3.2: Schematic representation for coordinate setting.

And if we were to write a line integral along this line, it would look like Equation 3.2, the Radon transform of function f(x, y):

$$P_{\theta}(t) = \int_{-\infty}^{\infty} f(x, y) \cdot \delta(x \cdot \cos(\theta) + y \cdot \sin(\theta) - t) dx dy$$
 (3.2)

Where δ , the delta function, is defined in Equation 3.3.

$$\delta(\phi) = \begin{cases} 1, & \phi = 0 \\ 0, & otherwise \end{cases}$$
 (3.3)

As I have mentioned previously, a projection is a set of line integrals such as $P_{\theta}(t)$. Geometry plays a very important role in how the integrals are written and solved for reconstruction. The simplest case is the one where the set is acquired in a row, describing what is called a parallel geometry. Another more complex case is when a single point source is used as origin for all rays, forming a fan. This is called a fanbeam array. There are other possible geometries, but they fall out of the scope of this work and will therefore not be addressed any further.

3.2.3 The Fourier Slice Theorem

The Fourier Slice Theorem (FST) is the most important component of the most important algorithm in tomographic inversion, the Filtered BackProjection algorithm (FBP). FST is based on the equality relation between the two-dimensional Fourier Transform (FT) of the object function and the one-dimensional FT of the object's projection at an

angle θ . Let's start by writing the 2D FT for the object function, Equation 3.4, and the 1D FT of projection P_{θ} , in Equation 3.5.

$$F(u,v) = \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} f(x,y) \cdot \exp\left[-j2\pi(ux+vy)\right] dxdy \tag{3.4}$$

$$S_{\theta}(\omega) = \int_{-\infty}^{\infty} P_{\theta} \cdot \exp\left[-j2\pi\omega t\right] \tag{3.5}$$

For simplicity, let's consider the 2D FT at the line defined by v=0 in the frequency domain. We rewrite the 2D FT integral as:

$$F(u,0) = \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} f(x,y) \cdot \exp\left[-j2\pi\omega ux\right] dxdy$$
 (3.6)

Notice that y is not present in the phase factor of the FT expression anymore, and this means we can rearrange the integral as:

$$F(u,0) = \int_{-\infty}^{\infty} \left[\int_{-\infty}^{\infty} \mathbf{f}(\mathbf{x}, \mathbf{y}) d\mathbf{y} \right] \cdot \exp\left[-j2\pi\omega u x \right] dx$$
 (3.7)

Now, the **bold** part of Equation 3.7 is similar to Equation 3.2. It is precisely that equation, considering $\theta = 0$ and a constant value of x, as in Equation 3.8.

$$P_{\theta=0}(x) = \int_{-\infty}^{\infty} f(x, y) dy$$
 (3.8)

This in turn can be substituted in Equation 3.7, finally arriving at:

$$F(u,0) = \int_{-\infty}^{\infty} P_{\theta=0}(x) \cdot \exp\left[-j2\pi ux\right] dx \tag{3.9}$$

And this is the one-dimensional FT for the projection at angle $\theta = 0$. Finally, the enunciation of the Fourier Slice Theorem:

The Fourier Transform of a parallel projection of an image f(x,y) taken at angle θ gives a slice of the two-dimensional Fourier Transform, F(u,v), subtending an angle θ with the u-axis (see Figure 3.3)

3.2.4 The Filtered BackProjection Algorithm

3.2.4.1 The rationale for FBP

If one takes the FST into account, the idea behind the FBP seems to appear almost naturally. Say one has a single projection and its Fourier transform. From the FST, this projection is the same as the object's two-dimensional FT in a single line. A crude reconstruction of the original object would result if someone were to place this projection in its right place in the Fourier domain and then perform a two-dimensional IFT, while assuming every other projection to be 0. The result, in the image space, would be as if someone had smeared the object in the projections direction.

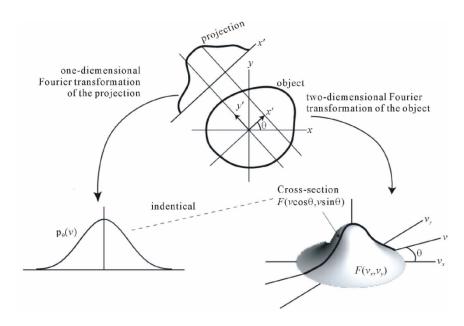


Figure 3.3: The FST, a schematic representation.

What is really needed for a correct reconstruction is to do this many times, with many projections. This brings a problem with the method: smearing the object in all directions will clearly produce a wrong accumulation in the center of the image, since every projection passes through the middle (remember we are still talking about parallel geometry projections) and are summed on top of each other, but on the outer edges, this does not occur. If one does not address this, the image intensity levels in the reconstructed image will be severely overestimated in the center and underestimated in the edges (due to normalization). The solution is conceptually easy: we multiply the Fourier transform by a weighting filter proportional to its frequency (ω) and that encompasses its relevance in the global scheme of projections. If there are K projections, then it is adequate for this value to be $\frac{2\pi|\omega|}{K}$. As an algorithm, FBP can be written as in Algorithm 1.

Algorithm 1 The Filtered BackProjection Algorithm

```
\begin{array}{l} \textbf{for all } \theta, \theta \in \left\{0..180, \frac{180}{K}\right\} \textbf{do} \\ \text{Measure projection } P_{\theta}(t); \\ \text{FT}(P_{\theta}(t)), \text{ rendering } S_{\theta}(\omega) \\ \text{Multiply by } \frac{2\pi|\omega|}{K}; \\ \text{Sum the IFT of the result in the image space.} \\ \textbf{end for} \end{array}
```

3.2.4.2 Fan Projections Reconstruction

Parallel projections, in which the object is scanned linearly from multiple directions, have the advantage of having a relatively simple reconstruction scheme. However, they

usually result in acquisition times which are in the order of minutes. A faster way of collecting the data is one where all radiation emanates from a single point-source, which rotates around the target object (as well as the detectors). There are two types of fan beam projections: equiangular and equally spaced. In this project, I have only worked with equiangular processes, so I will not include an explanation for equally spaced fan beam projections. The reader may find this well described (much better than I would be able to) in [9] and [8].

Consider Figure 3.4. If our projection data were acquired through a parallel ray geometry, we would be able to say that ray SA belonged to a projection $P_{\theta}(t)$, in which θ and t would be written:

$$\theta = \beta + \gamma$$
 and $t = D \cdot \sin \gamma$ (3.10)

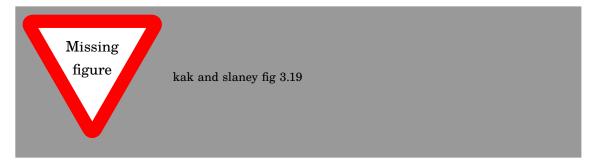


Figure 3.4: Schematic representation of an equiangular fan beam projection.

3.3 DOAS

3.4 DOAS tomography

3.4.1 Active DOAS tomography

3.4.2 Passive DOAS tomography

 $_{\text{Chapter}}$ 4

Research Methodology

- 4.1 Aimed contribution
- 4.2 Detailed work plan and scheduling
- 4.3 Validation methodology
- 4.4 Integration with other research activities

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