Title

Science Requirements Document for SPEXone

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DOCUMENT CHANGE RECORD

Issue	Date	Changed Section	Description of Change
1.0	23-11-2017		First issue
1.1	15-11-2018	Section 6 and Appendix A	Changed definition of spatial resolution in footnote of Table 1. Better in line with Gauss now. Also changed minimum LER spectrum in Appendix A for longer wavelengths. Now the values are representative for the angle average instead of angle with minimum radiance.
1.2	12-3-2019	Section 6	Modified sampling requirement to factor 1.5-2 over-sampling instead of fixed value of 2.

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1 INTRODUCTION

This document summarizes the science requirements and corresponding high-level instrument requirements for the SPEXone polarimeter for the NASA Phytoplankton, Aerosols, Clouds, and ocean Ecosystems (PACE) mission. The launch of PACE is planned August 2022. PACE is an observatory to include three instruments: as a primary instrument the Ocean Color Instrument (OCI), with the goal to understand and quantify global biogeochemical cycling and ecosystem function in response to anthropogenic and natural environmental variability and change. The mission also comprises two polarimeters: SPEXone with its unprecedented polarimetric accuracy focusses on the characterization aerosols (e.g. absorption, composition, size, height) and the wide-swath hyper-angular HARP polarimeter instrument focusses on the characterization of clouds. The primary goal of the Dutch SPEXone instrument is to understand and quantify the role of aerosols in the climate system. Moreover, SPEXone in combination with HARP and OCI will provide new insight in how aerosols are influencing the formation of clouds. An additional goal of the SPEXone instrument is to provide an atmospheric correction (aerosols and clouds) to facilitate ocean colour retrievals from OCI. It must be noted that due to the very high accuracy but limited swath SPEXone can act as a research demonstrator/enabler for a future breakthrough air quality instrument to improve air quality forecasts of Particulate Matter (PM).

The added value of SPEXone to the PACE mission is thus:

- SPEXone with its unprecedented polarimetric accuracy provides accurate characterization of aerosols for climate and air-quality applications. The wide-swath hyper-angular HARP polarimeter instrument focusses on the characterization of clouds. The HARP polarimeter provides similar information as the ASPIM module that was earlier proposed by the NL consortium in addition to SPEX but was descoped because of budgetary reasons. In essence, HARP compensates for this descope.
- SPEXone provides the necessary data for enhanced light path correction for the Ocean Color Instrument (OCI) measurements, which significantly enhance the OCI science. The OCI instrument focusses on better understanding of the ocean biosphere response to anthropogenic and natural environmental variability and change, and provides missing information to better manage fisheries and responses to phytoplankton blooms.

Anthropogenic aerosols are believed to cause a forcing of climate change comparable in magnitude but opposite in sign to greenhouse gases. In contrast to the climate effect of greenhouse gases, which is understood relatively well, the negative forcing (cooling effect) caused by aerosols represents the largest reported uncertainty in the most recent assessment of the International Panel on Climate Change (IPCC). This uncertainty severely hampers future predictions of climate change. Strong aerosol cooling in the past and present would imply that future global warming may proceed at, or even, above the upper extreme of the range projected by the IPCC. SPEXone could then fulfil the “A” in PACE and in combination with HARP and OCI on the PACE observatory allow for a breakthrough in climate research.
Aerosols are also known to strongly affect air quality, especially in regions with high industrial activity and large amounts of traffic, or in regions that are influenced by biomass burning. Exposure to particulate matter air pollution has major adverse human health impacts, including asthma attacks, heart and lung diseases, and premature mortality. Due to the very high accuracy but limited swath SPEXone can act as a research demonstrator/enabler for a future breakthrough air quality instrument.

The requirements for the SPEXone polarimeter are driven by the most important science questions related to the role of aerosols in climate and air quality. These science questions, and the flow down to level-2 requirements (i.e. on geophysical products) are based on various contributions in the scientific literature [e.g. /*Mishchenko et al., 2004*/, /Loeb and Su, 2010/, /*Fridlind and Ackerman, 2011*/], supported and confirmed by the latest (5th)

Assesment Report (AR-5) of the Intergovernmental Panel on Climate Change (IPCC), the Global Climate Observing System (GCOS) of the World Meterological Organization (WMO), the studies for the NASA Aerosol and Cloud Experiment (ACE), and the PACE-SPEX user workshop held 27 November 2015 at NWO in the Hague with representatives of the most important Dutch user groups and international experts from the aerosol research community. The flow down from level-2 requirements to level-1 requirements is based on extensive retrieval simulations performed at SRON, Netherlands Institute for Space Research [/Hasekamp and Landgraf, 2007/; /Hasekamp 2010/; /Wu et al., 2015/, /Wu et al., 2016/].

Section 2 summarizes the scientific background and objectives, section 3 discusses observational needs for climate and air quality applications, section 4 summarizes current aerosol remote sensing capabilities, section 5 and 6 provide the level-2 and level-1 requirements, respectively. Finally, section 7 summarizes the different requirements and relates them to each other.

2 SCIENTIFIC BACKGROUND

2.1 Aerosols and Climate Sensitivity

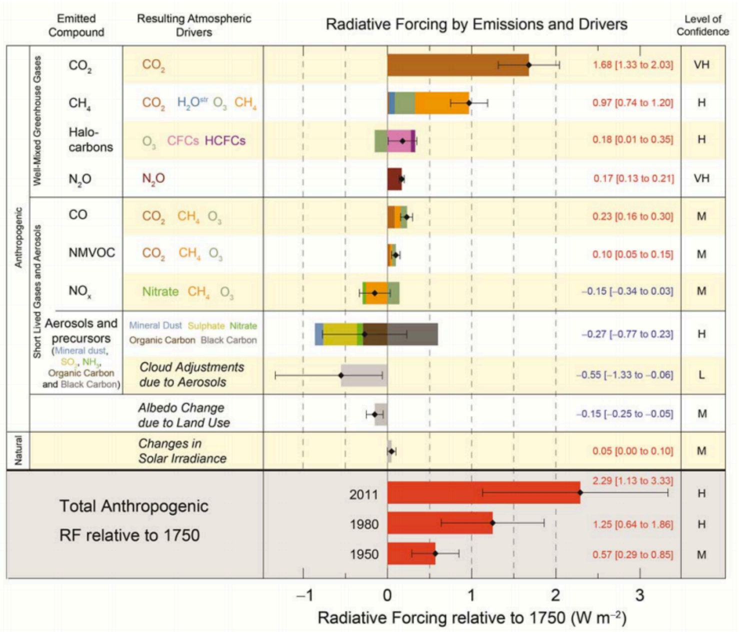

Figure 2-1. Magnitude and error bars of different radiative forcing terms as given in the IPCC Assessment Report-5 (AR5)

Aerosols represent the largest uncertainty in climate research. This uncertainty severely hampers our ability to model the effect of increasing greenhouse gas concentrations on the global temperature. The temperature change ΔT as a result from doubling the atmospheric CO_2 concentration is often referred to as the 'climate sensitivity'. Current estimates of climate sensitivities vary widely in a range from 2.0-4.5 K (/*Knutti and Hegerl, 2008*/), and even higher values cannot be ruled out. The uncertainty in estimates of the climate sensitivity did hardly improve ever since the first estimates were made. For example, /*Arrhenius, 1896*/ and /*Callendar, 1938*/, estimated values for the climate sensitivity of 5.5K and 2K, respectively. These estimates are still very close to the upper and lower bounds of current estimates.

In order to estimate climate sensitivity one needs to know how the change of the global temperature a result of a certain change in radiative forcing ΔF. This quantity is often referred to as the climate sensitivity parameter λ:

$$\lambda = \Delta T \;/\; \Delta F.$$

The climate sensitivity parameter can be estimated from the global temperature record over a given time period if the change in radiative forcing ΔF is known over the same period. ΔF is roughly described by the sum of the contribution of greenhouse gases ΔF_{ghg} and the contribution of aerosols ΔF_{aer}.

$$\Delta F = \Delta F_{ghg} + \Delta F_{aer}.$$

Here, the contribution of greenhouse gases ΔF_{ghg} over the industrial period is about 2.6 Wm^{-2} and is known relatively accurate as indicated by the Intergovernmental Panel on Climate Change (IPCC, see Figure 2-1). However, the contribution of aerosols is largely uncertain and the IPCC indicates a range from about +0.5 Wm-2 to -2.0 Wm^{-2}. So, the uncertainty in aerosol radiative forcing poses a severe limitation to estimate the climate sensitivity. This means that the historical temperature record can be equally well reproduced by a climate model with small amount of aerosol cooling, and a small climate sensitivity, or by a climate model with a large amount of aerosol cooling, and a large climate sensitivity. Two such model simulations are shown respectively by the blue and red line in Figure 2-2 (adopted from /*Knutti and Hegerl, 2008*/). Despite the fact that these two models reproduce the historical temperature record equally well, they do give a very different prediction for the temperature change in the 21st century, with difference between ~2.5 K and ~6 K for the year 2100. Clearly, a better understanding of the aerosol radiative forcing is essential in order to obtain reliable predictions for future temperature change.

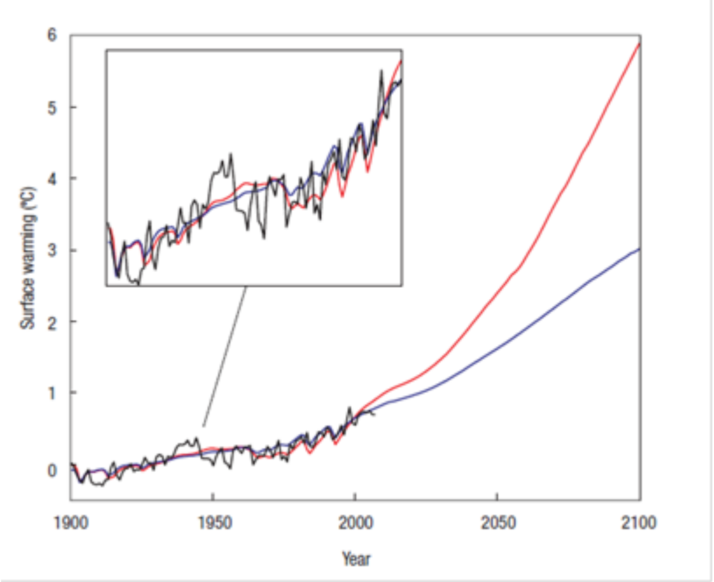

Figure 2-2 (adopted from /Knutti and Hegerl, 2008/): Global temperature change since the year 1900 as simulated by a climate model with small amount of aerosol cooling, and a small climate sensitivity (blue line), and by a climate model with a large amount of aerosol cooling and a large climate sensitivity (red line).

2.1.1 Radiative Forcing by Aerosols

Aerosols affect climate directly by scattering and absorption of solar radiation, and by scattering, absorption, and emission of thermal radiation. The corresponding radiative forcing is indicated as the direct aerosol radiative forcing. Aerosols also affect the climate indirectly by changing the macro-physical and microphysical properties of clouds (indirect and semi-direct effect).

The Direct Aerosol Effect

In the latest (5th) assessment report of the IPCC the direct aerosol radiative forcing is estimated to be -0.3 Wm^{-2} with an uncertainty of 0.5 Wm^{-2} at the 90% confidence level. This is an increase in uncertainty of 0.1 Wm^{-2} compared to IPCC 4th assessment report (AR4) which seemed to suggest a better quantification of the direct forcing compared to AR-3.

The study of /*Loeb and Su, 2010*/ suggests that the uncertainty on the direct aerosol radiative forcing may be even factor 2-4 greater than the value cited in the latest IPCC assessment. /*Loeb and Su, 2010*/ obtained their results by analyzing the effect on the radiative forcing when perturbing aerosol properties such as Aerosol Optical Thickness (AOT), Single Scattering Albedo (SSA), asymmetry parameter of the scattering phase function, aerosol scale height, anthropogenic fraction) within realistic uncertainty bounds. Hence, their results are not dependent on the different assumptions made in climate models. Insufficient knowledge of the aerosol single scattering albedo was found to have the largest effect on the uncertainty on the direct aerosol radiative forcing.

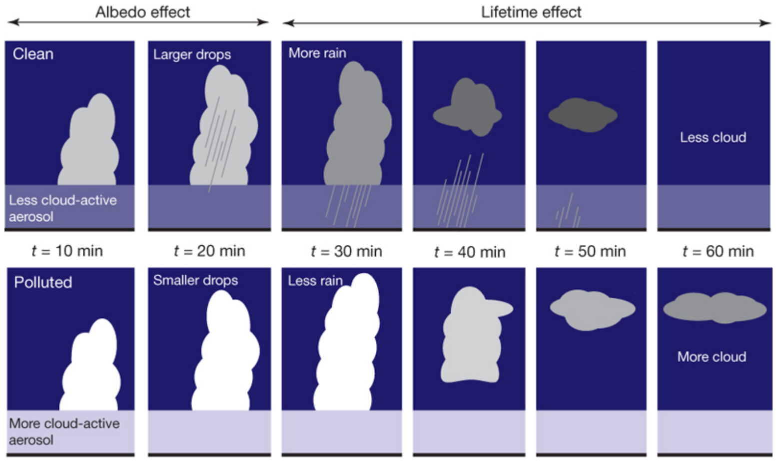

Figure 2-3 (from /Stevens and Feingold, 2009/): Schematic explanation of the first and second aerosol indirect effect .

Aerosol Effects on Clouds (Indirect and Semi-Direct effects)

Aerosols can affect cloud properties in various ways, as described in the review paper by */Lohmann and Feichter, 2005/*. A short summary will be given here. The first indirect effect, referred to as the "Twomey effect" or "cloud albedo effect", is the effect that an increase in aerosol concentration leads to a cloud with more but smaller cloud droplets, which results in a higher reflectivity of the cloud (*/Twomey, 1959/*). The second aerosol indirect effect is the effect that smaller cloud droplets (due to more aerosol particles competing for the same amount of water vapor) decrease the precipitation efficiency which results in a prolonged cloud lifetime. For a schematic explanation of the first and second aerosol indirect effect see Figure 2-3 (adopted from */Stevens and Feingold, 2009/*). The semi-direct aerosol effect is the effect that absorption of solar radiation by soot particles or mineral dust warms the atmospheric aerosol layer, which could hinder cloud formation or cause cloud droplets to evaporate [*/Lohmann and Feichter, 2001/*; */Penner et al, 2003/*; */Koren et al, 2004/*]. Aerosols can also affect mixed-phase clouds through the thermodynamic effect (smaller droplets leads to delay in freezing) and the Glaciation indirect effect (more ice nuclei increase the precipitation efficiency). For more details we refer to the paper of */Lohman and Feichter, 2005/*, and references therein. Clouds are an important regulator of the Earth's radiation budget. About 60% of the Earth surface is covered with clouds. On global average, clouds cool the Earth-atmosphere system, as the energy loss of 48 Wm^{-2} (by reflection back to space) is only partially compensated (30 Wm^{-2}) by trapped infrared radiation by clouds. Small changes in macrophysical and microphysical properties can have a significant effect on climate. For example, a 5% increase of the shortwave cloud forcing would compensate the increase in greenhouse gases in the period 1750-2000 [/*Ramaswamy et al., 2001*/]. Hence, it is extremely important to understand and quantify the effects of aerosols on cloud properties.

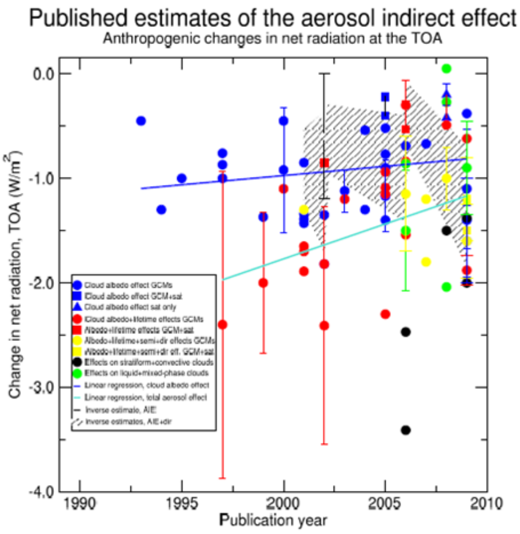

Figure 2-4: Estimates of the indirect aerosol radiative effect as function of publication year. The figure is adopted from /*Lohmann et al., 2010*/

The overall aerosol effect related to clouds is generally considered to be larger and even less well understood than the aerosol direct effect. In the latest IPCC assessment report the radiative forcing due to the aerosol indirect effect is estimated to be between -0.06 and -1.33 Wm^{-2} with a median value of -0.55 Wm^{-2}. /*Lohmann et al.,2010*/ nicely summarized different estimates of the indirect aerosol radiative forcing as a function of publication year (see Figure 2-4). From this figure it follows that estimates of the first indirect effect range between -0.2 and -1.5 Wm^{-2} whereas the estimates that also include the cloud lifetime effect range between about +0.1 and -2.0 Wm^{-2}. The range between the different estimates of the indirect aerosol effect, which is a measure for the uncertainty, did hardly narrow from the early 1990s till present. Most of the estimates shown in Figure 2-4 [/*Lohmann et al., 2010*/] are based on simulations of Global Circulation Models (GCMs) but some estimates include empirical relationships between aerosols and cloud properties from satellite data or are solely based on satellite data. The estimates that are (partly) based on satellites yield smaller negative forcing (less cooling) than the GCM estimates, but it should be noted that aerosol-cloud relationships from currently available satellite products give rise to numerous problems (see below).

In the past decades, our understanding of aerosols and their potential effects on clouds and climate has been significantly improved. However, this did not lead to a reduced uncertainty on quantitative estimates of the total aerosol radiative forcing. In fact, intensified aerosol research after the 4th assessment report of the IPCC did lead to the insight that the actual claimed uncertainties on the total aerosol direct radiative forcing are even larger than those reported earlier by the IPCC. In the latest IPCC assesment (AR5) even a total positive aerosol forcing cannot be excluded anymore.

2.2 Aerosols and Air Quality

Aerosol, in the context of air quality often referred to as 'particulate matter', is one of the key pollutants affecting human health. The smallest particles can penetrate deep into our respiratory systems, and consequently present a significant health-risk. Ground based measurement networks are set up worldwide to measure aerosol for air quality applications. Despite their abundance, such networks by construction provide only local information. Typically, ground based aerosol data classify air-quality by means of aerosol size as PM10 and PM2.5, while their chemical composition can be achieved only by elaborate chemical analyses on a even more local scale. Aerosol measurements obtained by satellite remote sensing have the potential to significantly contribute to air quality research. These measurements, assimilated into atmospheric chemistry transport models, have the potential capability to quantify aerosol sources, which is very important for air quality forecasts. Currently, data assimilation of satellite aerosols data is almost exclusively restricted to the aerosol optical thickness. With these data, aerosol sources can only be poorly quantified, as different combinations of various aerosol types (originating from different sources) will lead to the same optical thickness. The key information that is missing to make a significant advance in data assimilation of aerosols is the identification of aerosol type, which is an important challenge in aerosol satellite remote sensing. This aspect is only to limited extend covered by current and planned satellite instruments.

2.3 SpexOne Science Questions

The Science questions that SpexOne aims to address can be summarized as follows:
How large is the Direct Radiative Effect of Aerosols (DREA)?

- What is the anthropogenic contribution to the DREA?
- What is the contribution of different sources to the DREA?
- What is the difference of the DREA contribution for cloudy and clear skies.

How large is the indirect effects of aerosols?

- What is the relationship between aerosol and cloud properties?
- How does the relationship depend on aerosol type?
- How do absorbing aerosols affect cloud evaporation?

How do aerosols affect air quality?

- What is the importance of different aerosol emissions?

3 AEROSOL OBSERVATIONAL NEEDS FOR CLIMATE AND AIR QUALITY

3.1 Climate applications: Direct Radiative Effects

The direct aerosol effect refers to the effect of aerosols on climate through their interaction with solar radiation. Aerosols that mainly scatter sunlight have a cooling effect on climate, while aerosols that absorb light mainly warm the aerosol layer in the atmosphere. To determine the impact of aerosols on climate it is essential to know, in addition to the Aerosol Optical Thickness (AOT) and the scattering phase function, to which extend aerosols absorb solar radiation. Hereto, it is necessary to measure in addition the Single Scattering Albedo (SSA) or Aerosol Absorption Optical Thickness (AAOT). Furthermore, the direct radiative effect of (either absorbing and/or scattering) aerosols depends on the albedo of the underlying surface or cloud, while scattering aerosols are mainly effective as climate forcing in clear skies. So, for the direct aerosol radiative effect auxiliary information on surface reflection and cloud properties is needed in addition to aerosol properties above clouds. Finally, the aerosol direct radiative effect depends on the aerosol layer height, especially for cloudy situations.

An alternative way to quantify the aerosol direct radiative effect above clouds is by calculating the difference between the total outgoing irradiance and the outgoing irradiance for an atmosphere with only clouds. Here, the total outgoing irradiance can be estimated from the measured spectral radiance. Currently, this is being done from single viewing angle radiance measurements but this estimate can be significantly improved using multi-angle measurements. To best estimate the outgoing irradiance combined measurements of cloud and aerosol properties are needed.

To summarize, the following observational needs exist for an improved **quantification of the direct aerosol effect**:

- Spectrally Resolved Aerosol Optical Thickness (AOT)
- Spectrally Resolved Single Scattering Albedo (SSA)
- Aerosol Phase Function
- Aerosol Layer Height
- Cloud Properties (Optical Thickness, effective radius, height, fraction)
- Surface Bidirectional Reflection Distribution Function (BRDF)
- Multidirectional Radiance Measurements from the UV to the SWIR.

For a better understanding of aerosol-climate interactions (e.g. dust, sea salt, biogenic aerosols) and determination of the impact of various aerosol types of different anthropogenic origin (e.g. industrial, biomass burning), it is important to relate the direct aerosol radiative effects to specific aerosol sources. For this purpose additional information on aerosol type / chemical composition (a.o through complex refractive index and particle shape) is needed. Only through such aerosol type characterization the anthropogenic contribution to the direct climate effects can be derived.

3.2 Climate Applications: Aerosol Effects on Clouds

To quantify the indirect aerosol effects a good proxy for the concentrations of Cloud Condensation Nuclei (CCN) at cloud base is needed. In many current studies the AOT is used as proxy for CCN at cloud base but this is not sufficient. To improve on this, information on aerosol layer height, aerosol type, and aerosol size distribution is needed, preferably for aerosol particles in 'dry' form (i.e. without water). From the cloud perspective information on cloud droplet size distribution and optical thickness are required. It would be extremely valuable if the cloud droplet concentration can be derived without adiabatic assumptions. Additionally cloud droplet size distribution and optical thickness would be needed. For ice clouds ice crystal concentration, crystal size, and crystal shape are required, and to determine for the semi-direct effect also information on aerosol absorption is needed.

The following observational needs exist to better **quantify the aerosol effects on clouds**:
- Aerosol Height
- Aerosol Amount
- Aerosol Type
- (dry) Aerosol Size Distribution.
- SSA or AAOT.
- Cloud Droplet Concentration
- Cloud Droplet Size
- Ice Crystal Concentration
- Ice Crystal Size
- Ice Crystal Shape
- Cloud Optical Thickness

For the indirect aerosol climate forcing and anthropogenic contribution to the observed aerosol changes and variability it is important to relate the indirect aerosol radiative effect to specific aerosol sources. For this purpose information on aerosol type / chemical composition (a.o through complex refractive index and particle shape) is needed.

3.3 Aerosol modeling / Emissions / Air Quality

Current comparisons between global Chemistry Transport Models (CTMs) / Climate Models and aerosol satellite observations are mostly limited to the AOT. Here, good agreement can be obtained despite still limited modeling of the exact regionally varying aerosol composition. The aerosol complex refraction index gives important additional information for model evaluation and provides insight in how well a model can simulate the aerosol composition, including Inorganics (Sulfate, Nitrate, and Ammonium), Organic Carbon, Black Carbon, Dust, Sea Salt, and water content.

The sensitivity of refractive index to aerosol composition is also expected to be very useful for air quality applications, such as monitoring and forecasting of air quality. In the Netherlands (TNO, RIVM, KNMI) the LOTOS-EUROS model is used for assimilating satellite data for air quality applications. The assimilation of satellite data to improve air quality analysis and forecasts is already very mature for trace gases such as NO2. However, assimilating aerosols (PM - Particulate Matter) is still a very big challenge. Currently, assimilation systems such as LOTOS-EUROS make use of satellite measurements of the AOT. Although this obviously improves the modeled AOT fields, modeled PM fields so far are hardly improved. Also, source apportionment is very limited when only AOT fields are assimilated. To improve this, information on aerosol type/composition is needed. Furthermore, information on the aerosol size distribution is needed because the finest particles are most harmful for human health. Furthermore, information on aerosol layer height is required for air quality.

Another important topic is aerosol emissions by biomass burning. Aerosol emissions caused by fires are very uncertain, witnessing the factor of 3 mismatch between bottom-up and top-down estimates. Global scale information on the ratio between Organic Carbon (OC) and Black Carbon (BC) will help in better characterizing fires (smoldering / flaming) and will allow to quantify their impact on climate through aerosol radiative effects. Information on aerosol height is also important for the climate effects of biomass burning aerosols.

The following observational needs exist to improve data assimilation for aerosol air quality monitoring and forecast
- Aerosol amount.
- Complex refractive index (aerosol type/composition), also in relation to characterize biomass burning emissions
- Aerosol size distribution
- Aerosol layer height

4 CURRENT AEROSOL REMOTE SENSING CAPABILITIES

The majority of satellite instruments used for aerosol retrieval are passive instruments, which means that they rely on the sun as a light source. An exception is the CALIOP instrument /*Winker et al., 2010*/ which is a LIDAR that provides accurate information on the vertical distribution of aerosols and clouds at high resolution.
From the passive satellite instruments used for aerosol retrieval, most are based on multi-wavelength, single-viewing angle observations of the intensity of light reflected from the Earth atmosphere and surface. With this type of instruments it is possible to derive the Aerosol Optical Thickness (AOT) and some semi-quantitative aerosol microphysical properties, such as Angstrom exponent (wavelength dependence of AOT as proxy for size) and fine mode fraction, but the retrievals rely heavily on the assumed microphysical aerosol model /*Kokhanovsky et al., 2010*/. Retrievals are much less reliable over land than over the dark ocean, because over land accurate a priori information on surface reflection is needed. The most widely used single viewing angle intensity instrument is the MODerate resolution Imaging Spectroradiometer (MODIS) which flies on both the NASA satellites Terra and Aqua. Other examples are the Medium Resolution Imaging Spectrometer (MERIS) on ENVISAT, the Advanced Very High Resolution Radiometer (AVHRR) on METOP (but a long record exists dating back to 1981 on various platforms), and the Ozone Monitoring Instrument (OMI) on Aura (NASA).

The next category of instruments measures the intensity in multiple viewing geometries of one ground scene, such as the Multiangle Imaging SpectroRadiometer (MISR) on the NASA Terra satellite. The multi-viewing capability helps in separating atmospheric scattering from surface reflection and also allows one to distinguish two to four compositional groups based on refractive index, and four size groups in the range of 0.1–2.0 micron (/*Kahn et al., 1998*/). The Advanced Along Track Scattering Radiometer (AATSR) observes one ground scene under 2 viewing angles and hence also falls in this class of instruments.

Aerosol properties can only be unambiguously determined from instruments that measure both intensity and polarization at multiple viewing angles for one scene (/*Mishchenko and Travis, 1997*/, /*Hasekamp and Landgraf, 2007*/). The only instrument of this type that has provided a multi-year data record is the POLarization and Directionality of Earth Reflectances-3 (POLDER-3) instrument on the PARASOL microsatellite, in orbit between 2004-2013. The retrieval algorithms used for the operational aerosol data products of POLDER do not make full use of the information contained in the measurements. However, the recent studies by /*Dubovik et al., 2011*/, /*Hasekamp et al., 2011*/ and /*Tanre et al., 2011*/ do fully exploit this information, and provide insight in the capabilities and limitations of the POLDER instrument, which has the highest aerosol retrieval capability of all (passive) instruments currently in space. The Aerosol Polarimetry Sensor (APS), with higher polarimetric accuracy, more viewing angles, and an extended spectral range compared to POLDER, unfortunately failed for launch in March 2011.

In the past years, satellite observations have played an increasingly important role in attempts to quantify the direct and indirect anthropogenic aerosol forcings on climate. For example, efforts have been made to use (mostly) MODIS satellite observations to estimate Direct Aerosol Radiative Effect (DREA) [e.g. /*Bellouin et al, 2005; 2008*/; /*Kaufmann et al, 2005*/; /*Chung et al., 2005*/; /*Remer et al, 2009*/]. A limitation of these studies is that the Single Scattering Absorption (SSA), indicative for aerosol absorption, is being taken from aerosol models. These models are known to be very inaccurate in SSA (/*Lacagnina et al;., 2015*/) while accurate SSA measurements are of crucial importance to DREA estimates /*Loeb and Su, 2010*/. In a first attempt to come with an improved DREA estimate, /*Lacagnina et al, 2017*/ used newly available information on SSA from the PARASOL satellite. Although this first DREA estimate based on SSA observations shows significant differences with earlier observational based estimates, the authors conclude that more accurate SSA information is needed to draw definite conclusions. Indeed, PARASOL SSA accuracy is about 0.05 which is a factor ~2 higher than requirements formulated within the NASA ACE study /ACE/ (https://acemission.gsfc.nasa.gov/documents/ACE_Report5_Aerosol_Science_v7.pdf) and the Global Climate Observing System /GCOS/ (https://www.ncdc.noaa.gov/gosic/gcos-essential-climate-variable-ecv-data-access-matrix/gcos-atmosphere-composition-ecv-aerosols-properties).

Also, many observational studies have been published looking at the cloud albedo effect (1st indirect effect) using e.g. MODIS (e.g. /*Kaufman et al, 2005*/, /*Quaas et al, 2008*/), ATSR-2 (e.g. /*Bulgin et al., 2008*/) or POLDER (e.g. /*Breon et al., 2002*/). These studies mostly point to a positive correlation between AOT and effective cloud droplet number concentration and a negative correlation between AOT and cloud droplet effective radius. Only for ATSR-2 observations the latter correlation was found to be positive in some cases. Moreover, numerous studies have been published indicating that satellite data show a positive correlation between AOT and Total Cloud Cover (TCC) (/*Sekiguchi et al., 2003*/, /*Loeb and Manalo-Smith, 2005*/, /*Kaufman et al, 2005*/, /*Matheson et al., 2006*/, /*Kaufman and Koren, 2006*/, /*Menon et al, 2008*/, /*Quaas et al., 2008*/). Such a relationship could provide evidence for the aerosol cloud lifetime effect (2nd indirect effect), and interpreting the relationships provided by satellites would imply a 3% increase in global cloud cover due to anthropogenic aerosols (/*Kaufman and Koren, 2006*/). The corresponding negative forcing would be almost enough to compensate a doubling in CO_2 concentration (/*Slingo, 1992*/). However, there is an ongoing debate in the recent literature whether the satellite derived relationships between AOT and cloud cover are caused by the cloud-lifetime effect or rather by retrieval artifacts or appararent correlations. [e.g. /*Stevens and Feingold, 2009*/, /*Quaas et al., 2010*/]. Indeed, /*Gryspeerdt et al., 2016*/ conclude that for a substantial part AOT- cloud fraction relations can be explained by aerosol swelling in the vicinity of cloud due to enhanced humidity.

Overall, current estimates of the direct and indirect aerosol radiative forcing that are based on satellite observations suffer from a number of important limitations (see e.g. /*Stevens and Feingold, 2009*/, /*Quaas et al., 2010*/, /*Brandey and Stier, 2010*/). The most important of these limitations are summarized below:

- **Aerosol absorption**:

Current satellite products do not provide sufficient information on aerosol absorption. There have been some attempts to derive the aerosol SSA from MODIS using the "critical reflectance method [/*Kaufman, 1989*/, /*Zhu et al., 2011*/] but this method can only be applied to observations at a certain surface albedo and, even more important, the accuracy of the method is significantly worse than what would be needed to be used in direct aerosol radiative forcing studies. Retrieval of aerosol SSA from UV measurements of TOMS and OMI [/*Torres et al., 1998*/] are restricted to the UV spectral range only whereas information on a broader spectral range is needed for radiative forcing estimations. With PARASOL an important step is made in providing satellite derived SSA products (/*Hasekamp et al., 2011*/, /*Dubovik et al., 2011*/, /*Lacagnina et al., 2015; 2017*/) but still a factor ~2 improvement is needed in SSA accuracy to reach the requirements as formulated within the NASA ACE studies and GCOS. As noted above, accurate knowledge on the aerosol SSA is critical for aerosol direct effect calculations (/*Loeb and Su, 2010*/). Hence, improved measurements of SSA are needed.

- **Anthropogenic fraction**

In order to estimate the effect of anthropogenic aerosols on climate, it is essential to know the fraction of anthropogenic and natural aerosols. When estimating the fraction of anthropogenic aerosols from MODIS data, it is often assumed that all fine mode aerosols are anthropogenic. This is a very crude assumption that results in large uncertainties in the anthropogenic fraction [/*Levy et al., 2007*/, /*Anderson et al., 2005*/; /*Chu et al., 2005*/]. In order to determine the fraction of anthropogenic aerosols more accurately, information on aerosol type is needed in addition to accurate size distribution retrievals. Aerosol complex refractive index and sphericity are important indicator for aerosol type (e.g. /*Russel et al, 2014*/, /*Li et al., 2013*/, /*van Beelen et al., 2014*/). Current satellites do not provide this information with sufficient accuracy.

- **Aerosol swelling near clouds**

Most satellite based studies find a positive relationship between AOT and total cloud cover, which sometimes is considered as evidence of the 2nd aerosol indirect effect (cloud lifetime effect). However, a recent study by /*Quaas et al., 2010*/, /Gryspeerdt et al., 2016/ indicates that relationships between AOT and total cloud cover can to a large extend be explained by the fact that in areas close to clouds, where the relative humidity is high, aerosol size increases considerable due to water uptake (depending on aerosol type). This leads to an increase in AOT, while the aerosol number concentration remains the same. This effect rsults in an apparent relationship between aerosols and total cloud cover but is not due to the aerosol effects on clouds. In order to account for this effect the amount of water uptake needs to be quantified.

- **Model validation with AOT**

Models will remain an essential tool to predict future climate, and hence a correct representation of aerosol effects on clouds and climate in these models is of utmost importance. In order for models to quantify the present-day radiative, models must be able to accurately calculate aerosol distributions and the corresponding microphysical and optical properties. Till now, model performance evaluation with satellite observations has been mostly restricted to measurements of the AOT. However, the same AOT values can be generated by different models for quite different mixtures of aerosol species, and hence aerosol microphysical and absorption properties (/*Kinne et al, 2006*/, /*Lacagnina et al., 2014*/). Comparison of more detailed aerosol properties (optical and microphysical) is needed to evaluate and improve model performance.

- **Aerosol height**

To study the interaction of clouds and aerosols, the aerosol height distribution is an important property to be measured, because aerosols can only interact with clouds if they are at the same altitude level. With most current passive remote sensing instruments it is very difficult to estimate the height of an aerosol layer. With active sensors, such as CALIOP/CALIPSO and Earthcare (to be launched by ESA in 2019) aerosol height can be accurately estimated but on cost of very sparse spatial coverage.

In conclusion, aerosol measurements from space are extremely challenging due to the large number of parameters involved and because of the difficulty of distinguishing the aerosol signal from the land-surface signal. To advance our understanding of the aerosol effect on climate and air quality, a multi-instrument/mission approach is needed including both active and passive sensors. For the passive sensors, there is growing consensus that multi-angle measurements of intensity and polarization are inevitable. The 3MI instrument on the operational METOP-SG mission of EUMETSAT will resume the observations of the POLDER instruments that provided observations in 1995/1996, 1997/1998, and between 2004-2013. 3MI uses the same filter-wheel technology as the POLDER instruments allowing for large spatial coverage but with limited accuracy of the polarization measurements. Earthcare will provide accurate information on the aerosol vertical distribution as identified in e.g. the NASA /*ACE*/ studies and /*GCOS*/. In particular, within NASA-ACE high accuracy polarimetry is identified as an important tool to enhance our knowledge on atmospheric aerosols.

5 SPEXONE LEVEL-2 PRODUCTS

5.1 Qualitative description

Aerosol Optical Properties.
Global measurements of AOT, phase function, and especially SSA or AAOT are needed for quantification of the direct and semi-direct aerosol effects. SPEXone will provide these quantities with unprecedented accuracy. In particular the high accuracy measurements of SSA will allow significant advances in quantification and understanding of the aerosol direct radiative effect.

Aerosol Height
Aerosol height is an important property for all applications considered here (direct effect, indirect effect, air quality, modeling, fires). SPEXone will provide an aerosol layer height product with an accuracy that is unprecedented for a passive remote sensing instrument. This information can be derived from polarimetric measurements in the near-UV where elevated aerosol layers affect the measured polarization by (partly) shielding the Rayleigh scattering signal. Studies with the Research Scanning Polarimeter have demonstrated that this is a powerful tool for aerosol height retrieval /*Wu et al., 2016*/. The information can be further improved by combination with the multi-angle measurements in the O2 a-band from SPEXone. The ultimate goal for an aerosol mission would be to combine a SPEX instrument with a LIDAR which is foreseen for the NASA-ACE mission, of which PACE is the pre-cursor. For selected scenes synergetic retrievals from SpexOne and the EarthCare LIDAR will be possible.

Aerosol Composition / Type
Information on aerosol composition and type is very important for the different applications. SPEXone will provide information on aerosol composition / type mostly through the complex refractive index. From studies using refractive index data from ground based AERONET measurements it has been demonstrated that it is possible to distinguish between Mineral Dust, Black Carbon, Organic Carbon, Inorganic Aerosol, and Water. The most important limitations for these AERONET based studies is the accuracy of the inferred refractive index and the fact that AERONET does not provide refractive indices separately for the fine and coarse mode. SPEXone will improve on both aspects, which allows a more accurate retrieval of the different aerosol components (including water) and also to separate between Mineral Dust and Sea-Spray aerosol. For further distinction between different inorganic aerosols synergy with aerosol precursor gas measurements from the geo-stationary satellites (Sentinel-4, TEMPO, GEMS) is expected to be a powerful tool.

Aerosol Size
Information on aerosol size is important for aerosol indirect effect studies and air quality applications. For the aerosol indirect effect, the aerosol size distribution is needed to obtain a good proxy for CCN information, preferably for particles in dry form. SPEXone provides aerosol size distribution information with unprecedented remote sensing accuracy. The aerosol water amount can be estimated from the refractive index (see above), which allows to obtain also the size distribution of the dry aerosol particles. For air quality applications, SPEXone can derive effective radius for values down to 0.05-0.1 micron, which is expected to provide important information for PM10 or PM2.5 monitoring and forecasts.

Cloud Properties
For aerosol indirect effect studies, cloud properties need to be measured in addition to aerosol properties. For Cloud properties PACE depends on the combination of SPEXone, HARP, and OCI. OCI will provide information on cloud droplet effective radius, cloud optical thickness, and cloud fraction. HARP combined with SpexOne will

deliver cloud height, cloud geometrical thickness, phase (ice / liquid), ice crystal aspect ratio and roughness, and an independent measure of cloud optical thickness. For stratiform clouds, the cloud droplet concentration can be derived from the COT, droplet size distribution, and cloud pressure thickness. Cloud droplet concentration is considered a very important cloud property that presently cannot be obtained from passive remote sensing.

5.2 Level-2 Requirements

This section presents the level-2 geophysical data products and the corresponding accuracy requirements needed to address the science questions formulated above, are given in this section. These requirements are adopted from studies performed for the NASA /*ACE*/ studies and /*GCOS*/.

Given the expected budget being available, for SpexOne the choice has been made to focus on the accuracy requirements in the studies mentioned above rather than on coverage. The SpexOne instrument is complimentary to 3MI with focus on a very high polarimetric accuracy (order of magnitude better than 3MI), measurements in the near-UV (down to 385 nm), high spatial sampling, and many measurements per individual ground pixel. This allows to make the next step in aerosol characterization needed to improve our understanding of the aerosol-climate effect, but with a reduced spatial coverage (swath of ~100 km). This means that the focus of SpexOne is on the science questions related to climate (direct and indirect effect) for which a swath of 100 km is sufficient (/*Geogdzhayev et al., 2013*/). Combination with the other instruments on PACE (the HyperAngular Rainbob Polarimeter - HARP and the Ocean Color Instrument - OCI), which provide very detailed and accurate information on clouds, the climate related science questions can be adressed with unprecedented accuracy. For air quality, a swath of 100 km of SpexOne itself is not sufficient to be used in air-quality forecasting. However, the SpexOne data are still very useful for validation of models - leading to a better understanding of aerosol processes. Furthermore, SpexOne data will together with OCI and HARP data be assimilated in the GEOS-5 model of NASA-GSFC. The assimilated product will be of great value to air quality monitoring.

5.2.1 Requirements on aerosol properties

Requirement lv2-aerosol-01
The aerosol optical thickness (AOT) shall be measured with an accuracy better than the largest of 0.03 or 10%. This accuracy is needed to improve on current estimates of direct aerosol radiative forcing.

Requirement lv2-aerosol-02
The aerosol Single Scattering Albedo (SSA) shall be measured with an accuracy better than 0.02 (Goal)-0.03 (threshold). This accuracy is needed to improve on current estimates of direct aerosol radiative forcing.

Requirement lv2-aerosol-03
The accuracy of the aerosol effective radius of fine and coarse mode aerosols shall be measured with an accuracy 10%. This accuracy is needed for: 1) determining the fraction of aerosols capable of acting as Cloud Condensation Nucleus (CCN) /Rosenfeld, 2006/ 2) determining aerosol water uptake (together with refractive index)..

Requirement lv2-aerosol-04
The accuracy of the aerosol effective variance of fine and coarse mode aerosols shall be measured with an accuracy better than 50%.

Requirement lv2-aerosol-05
The accuracy of the real part of the refractive index of the fine and coarse mode shall be measured with an accuracy better than 0.02. This accuracy is needed for aerosol type identification and for determining aerosol water uptake.

Requirement lv2-aerosol-06
The accuracy of the imaginary part of the refractive index of the fine and coarse mode shall be measured with an accuracy which is the largest 0.001 or 15%. This accuracy is needed for aerosol type identification and for determining aerosol water uptake.

Requirement lv2-aerosol-07
The accuracy of the aerosol column (cm-2) of the fine and coarse mode shall be better than its order of magnitude. This accuracy is needed to identify aerosol cloud relationships (cloud albedo effect, cloud lifetime effect, semi-direct effect), direct aerosol forcing under cloudy conditions, and to characterize the radiative properties of the Twilight Zone.

Requirement lv2-aerosol-08
The accuracy of the aerosol layer height shall be measured with an accuracy better than 500 meter. This accuracy is needed to determine whether aerosols are located below or above a cloud and whether they are able to interact with clouds (ice or water)

5.2.2 Requirements on cloud properties

Requirement lv2-cloud-01
The Cloud Optical Thickness (COT) shall be measured with an accuracy better than the largest of 0.10 or 10%. This accuracy is needed to determine the cloud radiative properties, and the effect of aerosols thereon, with sufficient accuracy.

Requirement lv2-cloud-02
The cloud top and base height shall be determined with an accuracy of 100 meter. (TBD).

Requirement lv2-cloud-03
The cloud droplet effective radius shall be measured with an accuracy better than the largest of 0.10 micron or 10%. This accuracy is needed to monitor the effect of aerosols on cloud droplet size (indirect effect)

6 INSTRUMENT REQUIREMENTS

The level-2 requirements of section 5.2 result in a number of instrument and level-1 requirements that are given in this section. These requirements are based on a number of sensitivity studies that relate requirements on aerosol properties to measurement characteristics for a satellite instrument [/Hasekamp and Landgraf, 2007/, /Veefkind et al., 2008/, /Hasekamp, 2010/, /Wu et al., 2015;2016/].

Requirement lv1-01
The instrument shall perform multi-spectral, multi-viewing-angle measurements of both intensity as well as degree of linear polarization (DoLP) of light reflected by the Earth atmosphere and surface. As shown by e.g. /Mishchenko and Travis, 1997/, /Chowdhary et al. 2001/, /Hasekamp and Landgraf, 2007/, and /Hasekamp, 2010/ this is the only instrument concept that allows to determine all relevant aerosol properties unambiguously.

Requirement lv1-02
Intensity and DoLP shall be measured in the VIS/NIR in the range 385-770 nm. The lower wavelength limit is driven by the accurate characterization of fine mode aerosols, Aerosol Layer Height, and for the characterization of absorbing aerosols. Also, the short wavelength edge is important for atmospheric correction for ocean color remote sensing. The long wavelength limit is driven by the need to include the O2 a-band, and to derive sufficient information on aerosol size.

Requirement lv1-03
The intensity shall be measured with a spectral resolution of 4 nm in the continuum (i.e. outside the O2 a- band). For the VIS/NIR spectral range aerosol shows a relatively strong spectral signature. Also for ocean applications spectrally resolved measurements in the (near-)UV and blue have added value. For the O2 a-band a 2nm spectral resolution for the radiance measurements is needed in order to resolve the O2 a-band absorption structure.

Requirement lv1-04
The DoLP shall be measured with a spectral resolution of 15 nm (@385 nm) – 40 nm (@770 nm) . This spectral resolution is sufficient to resolve all continuum features that are sensitive to aerosol properties

Requirement lv1-05
The Degree of Linear Polarization (DoLP) shall be measured with an accuracy of 0.003 in DoLP . This accuracy is needed to achieve the required accuracy for refractive index and single scattering albedo [/Hasekamp and Landgraf, 2007/, and /Hasekamp, 2010/].

Requirement lv1-06
The intensity shall be measured with an **accuracy** better than 2%. This accuracy is needed to achieve the required accuracy in aerosol optical thickness and single scattering albedo [/Hasekamp and Landgraf, 2007/, and /Hasekamp, 2010/]. The **Signal to Noise Ratio** (SNR) shall be better than 300 for the reference spectrum with moderate AOT over a dark ocean and a Solar Zenith Angle (SZA) of 70°. This SNR is defined at the required spectral and spatial resolution of polarization measurements.

Requirement lv1-07
Multi-viewing-angle observations shall be performed in the range +-57° (threshold). These angles are defined at ground level.

Requirement lv1-08
The intensity and Degree of Linear Polarization for one ground pixel shall be measured at at least 5 viewing. A recent study has shown that 5 viewing angles are sufficient for aerosol characterization [Wu et al, 2015]. For cloud characterization more angles (>20) are required. These measurements will be provided by the HARP instrument on PACE.

Requirement lv1-09
The common swath width of all viewing angles of the instrument shall be 100 km or larger.

Requirement lv1-10
The **spatial resolution** shall be better than 25 km^2 in order to have as much as possible scenes that are cloud free (or have only small cloud contamination) or homogeneously cloudy. The meaning of this spatial resolution requirement is that 55-60% of the integrated energy (for homogeneous illumination) should originate from a given 25 km^2 ground scene, and 90-95% should originate from an 100 km^2 scene, and >99% from a 225 km^2 scene. The Point Spread Function (PSF) shall be monotonically-decreasing function peaking at the spatial pixel under consideration.

Requirement lv1-11
The aspect ratio of the along track and across track dimensions of a spatial resolution element that corresponds to a 25 km^2 footprint shall be in the range 0.5 – 2.

Requirement lv1-12
The **spatial sampling distance** shall be a factor 1.5(T)-2(G) smaller than the dimensions of the spatial resolution element, in both the along track and across track direction.
This ensures that regridding on a common grid with a grid size equal to the spatial resolution element is possible for all viewing angles. In the end, all viewing directions will need to be re-constructed on a common spatial grid that needs to meet the spatial resolution requirement.

Requirement lv1-13
The uncertainty in the pointing knowledge of all viewing directions of the instrument shall be smaller than 0.1 spatial sampling distance (3-sigma) in both the along track and across track direction.

Requirement lv1-14
The instrument shall meet its performance for all observations with solar zenith angles of 70° or smaller.

Table 1: Overview of high-level L1 requirements of the SPEXlite-PACE polarimeter.

Parameter	**VNIR**
swath (common for all angles)	100 km
angular range (**on ground**)	+/- 57°
# viewing angles	5
spectral range	385-770 nm
spectral resolution intensity	4 nm continuum 2 nm O2 a-band (755-770 nm)
spectral resolution DoLP	15 nm @385 nm 40 nm @770 nm
spatial resolution[1] (for all angles)	25 km^2
aspect ratio along track / across track	0.5 - 2
spatial sampling ratio	1.5 (T) - 2(G)
polarimetric accuracy	0.003
radiometric accuracy	2%
pointing accuracy (for all angles)	0.1 spatial sampling distance
Signal to Noise Ration (SNR) at required resolution, spatial sampling 25 km^2, SZA = 70°, LER values of Table 4 (Appendix A)	300
Dynamic range: minimum value	LER values of Table 4 (Appendix A) for SZA=70°
Dynamic range: maximum value[2]	LER of 1.1, SZA = 15°
science observation range	SZA < 70 °

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[1] Spatial resolution is defined as the area on Earth from which 55-60% of the integrated energy shall originate. 90-95% of the energy shall originate from an area that is 4 times as large (factor 2 in both spatial dimensions). >99% of the energy shall originate from an area that is 9 times as large (factor 3 in both spatial dimensions).

[2] Note that the actual value for the maximum signal may be significantly larger over sun-glint, so the instrument should 'survive' larger values.

7 RELATION BETWEEN SCIENCE AND INSTRUMENT REQUIREMENTS

Table 2 summarizes the aerosol and cloud properties that are needed for the different applications of SpexOne on PACE.

Table 2: Aerosol and cloud measurements that are needed for the different applications of SpexOne on PACE as follows from /GCOS/ and /ACE/ and references therein (a.o. /Mishchenko et al., 2004/, /Loeb and Su, 2010/). Cloud properties will be obtained from synergy SpexOne, HARP, OCI.

Application	**Direct Radiative Effect of Aerosols**	**Aerosol effect on clouds**	**Air Quality**
Aerosol properties	• Spectrally resolved AOT • Spectrally Resolved SSA • Aerosol Phase Function • Aerosol Layer Height • Complex refractive index (to distinguish contribution from different sources (incl anthropogenic)	• Aerosol Height • Aerosol Number column • Aerosol Type • (dry) Aerosol Size Distribution. • SSA or AAOT.	• Aerosol number column. • Complex refractive index • Aerosol size distribution • Aerosol layer height
Cloud properties	• Cloud optical thickness • Droplet effective radius • Top and base height • Cloud fraction	• Cloud Droplet Concentration • Cloud Droplet Size • Ice Crystal Concentration • Ice Crystal Size • Ice Crystal Shape • Cloud Optical Thickness • Cloud fraction.	

Table 3 summarizes the level-1 requirements and relates them to the different level-2 requirements that are driving them.

Table 3: Overview of the different level-1 requirements and the relation to the level-2 requirements that are driving them

level-1 requirement	**driving level-2 requirements**	**Motivation**
lv1-01 (multi-angle, spectro polarimetric measurements)	lv2-aerosol-01 - lv2-aerosol-09	important for all scientific objectives
lv1-02 (lower wavelength limit)	lv2-aerosol-02 lv2-aerosol-06 lv2-aerosol-07 (for small mode)	-fine mode aerosol properties - absorption - Aerosol Layer Height
lv1-03 (spectral resolution intensity)	lv2-aerosol-08 lv2-cloud-02	-Retrieval of aerosol height distribution -retrieval of cloud top/base height - Capture spectral variation in DoLP of aerosols and surface
lv1-04 (spectral resolution DolP)		Capture spectral variation in DoLP of aerosols and surface
lv1-05 (polarimetric accuracy)	lv2-aerosol-01 lv2-aerosol-02 lv2-aerosol-05 lv2-aerosol-09	- Refractive index retrieval for type identification and water uptake - Aerosol Layer Height retrieval - Single Scattering Albedo retrieval (direct radiative forcing) - Aerosol Optical Thickness retrieval (direct radiative forcing) - retrieval of aerosols close to and above clouds
lv1-06 (radiometric accuracy)	lv2-aerosol-01 lv2-aerosol-02	- Retrieval of optical thickness - Retrieval of single scattering albedo
lv1-07 (angular range)	lv2-aerosol-01 - lv2-aerosol-07 lv2-aerosol-08	The range of scattering angles affects all microphysical and optical aerosol properties. The scattering angle range 90-130 degrees is most important for aerosol retrieval. The range 135-145 degrees is important for aerosol retrieval in cloudy scenes and cloud retrievals.
lv1-08 (angular resolution)	lv2-aerosol-09	5 angles needed for aerosol characterization
lv1-10 (spatial resolution)		- Detecting aerosol gradients - As much as possible pixels with small/no cloud cover

8 REFERENCES

ACE - Aerosol Clouds, & Ecosystems, Study Report, https://acemission.gsfc.nasa.gov/documents/ACE_Report5_Aerosol_Science_v7.pdf

Albrecht, B. A.: Aerosols, Cloud Microphysics, and Fractional Cloudiness, Science, 245, 1227–1230, doi:10.1126/science.245.4923.1227, 1989.

Anderson, T.L., et al., 2005a. Testing the MODIS satellite retrieval of aerosol fine-mode fraction. *J. Geophys. Res.*, **110,** D18204, doi:10.1029/2005JD005978.

Arrhenius, S. On the influence of carbonic acid in the air upon the temperature of the ground. *Phil. Mag.* **41,** 237–276 (1896).

Alexandrov, M.D., B. Cairns, and M.I. Mishchenko, 2012: Rainbow Fourier transform. J. Quant. Spectrosc. Radiat. Transfer, 113, 2521-2535, doi:10.1016/j.jqsrt.2012.03.025.

Anderson, T.L., et al., 2005a. Testing the MODIS satellite retrieval of aerosol fine-mode fraction. J. Geophys. Res., 110, D18204, doi:10.1029/2005JD005978.

Andreae, M., Jones, C., and Cox, P.: Strong present-day aerosol cooling implies a hot future, Nature, 435, 1187–1190, 2005.

van Beelen et al. Atmos. Chem. Phys., 14, 5969–5987, 2014 www.atmos-chem-phys.net/14/5969/2014/ doi:10.5194/acp-14-5969-2014

Bellouin, N., O. Boucher, D. Tanré, and O. Dubovik, 2003: Aerosol absorption over the clear-sky oceans deduced from POLDER-1 and AERONET observations. Geophys. Res. Lett., 30(14), 1748, doi:10.1029/2003GL017121.

Bellouin, N., O. Boucher, J. Haywood, and M.S. Reddy, 2005: Global estimates of aerosol direct radiative forcing from satellite measurements.
Nature, 438, 1138–1141.

Breon, F., Tanre, D., and Generoso, S.: Aerosol Effect on Cloud Droplet Size Monitored from Satellite, Science, 295, 834–838, doi:10.1126/science.1066434, 2002.

Bulgin, C. E., Palmer, P. I., Thomas, G. E., Arnold, C. P. G., Campmany, E., Carboni, E., Grainger, R. G., Poulsen, C., Siddans, R., and Lawrence, B. N.: Regional and seasonal variations of the Twomey indirect effect as observed by the ATSR-2 satellite instrument, Geophys. Res. Lett., 35, L02811, doi:10.1029/2007GL031394, 2008.

Callendar, G. S. The artificial production of carbon dioxide and its influence on temperature. *Q. J. R. Meteorol. Soc.* **64,** 223–240 (1938).

Charlson, R. J., Ackerman, A. S., Bender, F. A.-M., Anderson, T. L., and Liu, Z.: On the climate forcing consequences of the albedo continuum between cloudy and clear air, Tellus 59B, 715–727, 2007.

Chu, D.A., et al., 2002: Validation of MODIS aerosol optical depth retrieval over land. Geophys. Res. Lett., 29(12), doi:10.1029/2001GL013205.

Chung, C.E., V. Ramanathan, D. Kim, and I.A. Podgorny, 2005: Global anthropogenic aerosol direct forcing derived from satellite and ground- based observations. J. Geophys. Res., 110, D24207, doi:10.1029/2005JD006356.

Dubovik, O., M. Herman, A. Holdak, T. Lapyonok, D. Tanré, J. L. Deuzé, F. Ducos, A. Sinyuk, and A. Lopatin, 2011: Statistically optimized inversion algorithm for enhanced retrieval of aerosol properties from spectral multi-angle polarimetric satellite observations. Atmos. Meas. Tech. 4, 975–1018.

Dusek, U., G. P. Frank, L. Hildebrandt, J. Curtius, J. Schneider, S. Walter, D. Chand, F. Drewnick, S. Hings, D. Jung, S. Borrmann and M. O. Andreae, Size Matters More Than Chemistry for Cloud-Nucleating Ability of Aerosol Particles, Science 2 June 2006: Vol. 312 no. 5778 pp. 1375-1378 DOI: 10.1126/science.1125261

Forster, P., and Coauthors, 2007: Changes in atmospheric constituents and in radiative forcing. In Solomon, S., et al., Eds., Climate Change 2007: The Physical Science Basis. Cambridge Univ. Press, Cambridge, UK, pp. 129–234.

Fridlind, A. M., and A. S. Ackerman, 2011: Estimating the sensitivity of radiative impacts of shallow, broken marine clouds to boundary layer aerosol size distribution parameter uncertainties for evaluation of satellite retrieval requirements. J. Atmos. Oceanic Technol. 28, 530–538.

Geogdzhayev et al. Statistical analysis of single-track instrument sampling in space-borne aerosol remote sensing for climate research, JQSRT, 121(2013)69–77

GCOS, Global Climate Observation System, Aerosol Requirements, https://www.ncdc.noaa.gov/gosic/gcos-essential-climate-variable-ecv-data-access-matrix/gcos-atmosphere-composition-ecv-aerosols-properties.

Grandey, B. S. and Stier, P.: A critical look at spatial scale choices in satellite-based aerosol indirect effect studies, Atmos. Chem. Phys., 10, 11459-11470, doi:10.5194/acp-10-11459-2010, 2010

van Harten, G., F. Snik, J. Rietjens, J. M. Smit, J. de Boer, R. Diamantopoulou, O. P. Hasekamp, D. M. Stam, C. U. Keller, E. C. Laan, A. L. Verlaan, W. A. Vliegenthart, R. ter Horst, R. Navarro, K. Wielinga, S. Hannemann, S. G. Moon and R. Voors, Prototyping for the Spectropolarimeter for Planetary EXploration (SPEX): calibration and sky measurements, Proc. SPIE 8160, 81600Z (2011); doi:10.1117/12.893741

Hasekamp, O. P., 2010: Capability of multi-viewing-angle photo-polarimetric measurements for the simultaneous retrieval of aerosol and cloud properties. Atmos. Meas. Tech. 3, 839–851.

Hasekamp, O. P., and J. Landgraf, 2005: Linearization of vector radiative transfer with respect to aerosol properties and its use in satellite remote sensing. J. Geophys. Res. 110, D04203.

Hasekamp, O., and J. Landgraf, 2007: Retrieval of aerosol properties over land surfaces: capabilities of multiple-viewing-angle intensity and polarization measurements. Appl. Opt. 46, 3332–3344.

Hasekamp, O., P. Litvinov, and A. Butz, 2011: Aerosol properties over the ocean from PARASOL multi-angle photopolarimetric measurements. J. Geophys. Res., doi:10.1029/ 2010JD015469.

R. Kahn, P. Banerjee, D. McDonald, and D. J. Diner, "Sensitivity of multiangle imaging to aerosol optical depth and to pure-particle size distribution and composition over ocean," J. Geophys. Res. 103, 32195–32238 (1998).

Kaufmann, Y. J., 1989: The atmospheric effect on remote sensing and its correction. Theory and Applications of Optical Remote Sensing, G. Asrar, Ed., John Wiley & Sons, 336–428.

Kaufmann Y.J., et al., 2005a: Aerosol anthropogenic component estimated from satellite data. Geophys. Res. Lett., 32, L17804, doi:10.1029/
2005GL023125.

Kaufmann, Y. J. and I. Koren: Smoke and pollution aerosol effect on cloud cover, Science, 313, 655–658, doi:10.1126/science.1126232, 2006.

Kinne, S., M. Schulz, C. Textor, S. Guibert, Y. Balkanski, S. E. Bauer, T. Berntsen, T. F. Berglen, O.Boucher, M. Chin, W. Collins, F. Dentener, T. Diehl, R. Easter, J. Feichter, D. Fillmore, S. Ghan,P. Ginoux, S. Gong, A. Grini, J. Hendricks, M. Herzog, L. Horowitz, I. Isaksen, T. Iversen, A.Kirkevåg, S. Kloster, D. Koch, J. E. Kristjansson, M. Krol, A. Lauer, J. F. Lamarque, G. Lesins, X. Liu, U. Lohmann, V. Montanaro, G. Myhre, J. Penner, G. Pitari, S. Reddy, O. Seland, P. Stier, T. Takemura, and X. Tie, 2006: An AeroCom initial assessment – optical properties in aerosol component modules of global models. Atmos. Chem. Phys. 6, 1815–1834.

Knutti, R. and Hegerl, G. C., The equilibrium sensitivity of the earth's temperature to radiation changes, Nature Geoscience ; 1 ; 735-743, 2008

Kokhanovsky, A. A., J. L. Deuzé, D. J. Diner, O. Dubovik, F. Ducos, C. Emde, M. J. Garay, R. G. Grainger, A. Heckel, M.Herman, I. L. Katsev, J. Keller, R. Levy, P. R. J. North, A. S. Prikhach, V. V. Rozanov, A. M. Sayer, Y. Ota, D. Tanré, G. E. Thoma and E. P. Zege, 2010: The inter-comparison of major satellite aerosol retrieval algorithms using simulated intensity and polarization characteristics of reflected light. Atmos. Meas. Tech. 3, 909–932.

Koren, I., Kaufman, Y., Remer, L., and Martins, J.: Measurements of the effect of Amazon smoke aerosol on inhibition
of cloud formation , Science, 303, 1342–1345, doi:10.1126/science.1089424, 2004.

Koren, I., Remer, L. A., Kaufman, Y. J., Rudich, Y., and Martins, J. V.: On the twilight zone between clouds and aerosols, Geophys. Res. Lett., 34, L08805, doi:10.1029/2007GL029253, 2007.

Lacagnina, C., O. P. Hasekamp, and O. Torres (2017), Direct radiative effect of aerosols based on PARASOL and OMI satellite observations, J. Geophys. Res. Atmos., 122, doi:10.1002/2016JD025706

Lacagnina, C., O. P. Hasekamp, H. Bian, G. Curci, G. Myhre, T. van Noije, M. Schulz, R. B. Skeie, T. Takemura, and K. Zhang (2015), Aerosol single-scattering albedo over the global oceans: Comparing PARASOL retrievals with AERONET, OMI, and AeroCom models estimates, J. Geophys. Res. Atmos., 120, 9814–9836, doi:10.1002/2015JD023501.

Levy, R.C., et al., 2003: Evaluation of the Moderate-Resolution Imaging Spectroradiometer (MODIS) retrievals of dust aerosol over the ocean during PRIDE. J. Geophys. Res., 108(D19), 8594, doi:10.1029/ 2002JD002460.

Li et al., Atmos. Chem. Phys., 13, 10171–10183, 2013 www.atmos-chem-phys.net/13/10171/2013/ doi:10.5194/acp-13-10171-2013

Loeb, N. G., and W. Su, 2010: Direct aerosol radiative forcing uncertainty based on a radiative perturbation analysis. J. Clim. 23, 5288–5293.

Loeb, N. G. and Manalo-Smith, N.: Top-of-atmosphere direct radiative effect of aerosols over global oceans from merged CERES and MODIS observations, J. Clim., 18, 3506, 2005.

Lohmann, U. and Feichter, J.: Can the direct and semi-direct aerosol effect compete with the indirect effect on a global scale?, Geophys. Res. Lett., 28, 159–161, 2001.

Lohmann, U., and J. Feichter, 2005: Global indirect aerosol effects: a review. Atmos. Chem. Phys. 5, 715–737.

Lohmann, U., and S. Ferrachat, 2010: Impact of parametric uncertainties on the present-day climate and on the anthropogenic aerosol effect. Atmos. Chem. Phys. 10, 11373–11383.

Lohmann, U., L. Rotstayn, T. Storelvmo, A. Jones, S. Menon, J. Quaas, A. Ekman, D. Koch, and R. Ruedy, 2010: Total aerosol effect: Radiative forcing or radiative flux perturbation. *Atmos. Chem. Phys.*, **10**, 3235-3246, doi:10.5194/acp-10-3235-2010.

Matheson, M. A., Coakley Jr., J. A., and Tahnk, W. R.: Multiyear Advanced Very High Resolution Radiometer observa- tions of summertime stratocumulus collocated with aerosols in the northeastern Atlantic, J. Geophys. Res., 111, D15206,
doi:10.1029/2005JD006890, 2006.

Menon, S., Del Genio, A. D., Kaufman, Y. J., Bennartz, R., Koch, D., Loeb, N., and Orlikowski, D.: Analyzing signatures of aerosol-cloud interactions from satellite retrievals and the GISS GCM to constrain the aerosol indirect effect, J. Geophys. Res., 113, D14S22, doi:10.1029/2007JD009442, 2008.

Mishchenko, M. I., and L. D. Travis, 1997: Satellite retrieval of aerosol properties over the ocean using polarization as well as intensity of reflected sunlight. J. Geophys. Res. 102, 16989–17013.

Mishchenko, M., B. Cairns, J. Hansen, L. Travis, R. Burg, Y. Kaufman, J. Vanderlei Martins, and E. Shettle, 2004: Monitoring of aerosol forcing of climate from space: analysis of measurement requirements. J. Quant. Spectrosc. Radiat. Transfer 88, 149–161.

Mishchenko, M. I., B. Cairns, G. Kopp, C. F. Schueler, B. A. Fafaul, J. E. Hansen, R. J. Hooker, T. Itchkawich, H. B. Maring, and L. D. Travis, 2007: Accurate monitoring of terrestrial aerosols and total solar irradiance: introducing the Glory Mission. Bull. Amer. Meteorol. Soc. 88, 677–691.

Nakajima, T., and M. D. King, 1990: Determination of the optical thickness and effective radius of clouds from reflected solar radiation measurements. Part I: Theory. J. Atmos. Sci. 6, 1878–1893.

Platnick, S., 2000: Vertical photon transport in cloud remote sensing problems. J. Geophys. Res. 105, 22919–22935.

Quaas, J., Boucher, O., Bellouin, N., and Kinne, S.: Satellite-based estimate of the direct and indirect aerosol climate forcing, J. Geophys.
Res., 113, D05204, doi:10.1029/2007JD008962, 2008.

Quaas, J., B. Stevens, P. Stier, and U. Lohmann, 2010: Interpreting the cloud cover – aerosol optical depth relationship found in satellite data using a general circulation model. Atmos. Chem. Phys. 10, 6129–6135.

Remer, L., and co-authors, 2009, executive summary. Atmospheric aerosol properties and climate impacts, Report by the U.S. Climate Change Science Program and the Subcommittee on Global Change Research, M. Chin, R.A. Kahn, and S.E. Schwartz, Eds., NASA, 1-8

Riedi, J., P. Goloub, and R. T. Marchand (2001), Comparison of POLDER cloud phase retrievals to active remote sensors measurements at the ARM SGP site, Geophys. Res. Lett., 28(11), 2185–2188, doi:10.1029/2000GL012758.

Philip B. Russell, Melo Kacenelenbogen, John M. Livingston, Otto P. Hasekamp, Sharon P. Burton, Gregory L. Schuster, Matthew S. Johnson, Kirk D. Knobelspiesse, Jens Redemann, S. Ramachandran, and Brent Holben 2014. A multiparameter aerosol classification method and its application to retrievals from spaceborne polarimetry J. Geophys. Res. Atmos., 119, 98389863, doi:10.1002/2013JD021411.

Schuster, G. L., O. Dubovik, B. N. Holben, and E. E. Clothiaux, 2005: Inferring black carbon content and specific absorption from Aerosol Robotic Network (AERONET) aerosol retrievals. J. Geophys. Res. 110, D10S17

Sekiguchi, M., Nakajima, T., Suzuki, K., Kawamoto, K., Higurashi, A., Rosenfeld, D., Sano, I., and Mukai, S.: A study of the directand indirect effects of aerosols using global satellite data sets of aerosol and cloud parameters, J. Geophys. Res., 108(D22), 4699,doi:10.1029/2002JD003359, 2003.

Frans Snik, Theodora Karalidi, and Christoph U. Keller, "Spectral modulation for full linear polarimetry," Appl. Opt. 48, 1337-1346 (2009)

S. Solomon, J. S. Daniel, R. R. Neely, J.-P. Vernier, E. G. Dutton, L. W. Thomason, The Persistently Variable “Background” Stratospheric Aerosol Layer and Global Climate Change, Science: Vol. 333 no. 6044 pp. 866-870, DOI: 10.1126/science.1206027, 2011

Stevens, B., and G. Feingold, 2009: Untangling aerosol effects on clouds and precipitation in a buffered system. Nature 461, 607–613

Tanré, D., F. M. Bréon, J. L. Deuzé, O. Dubovik, F. Ducos, P. François, P. Goloub, M. Herman, A. Lifermann, and F. Waquet, 2011: Remote sensing of aerosols by using polarized, directional and spectral measurements within the A-Train: the PARASOL mission. Atmos. Meas. Tech. 4, 1383–1395.

Torres, O., P. K. Bhartia, J. R. Herman, Z. Ahmad, and J. Gleason (1998), Derivation of aerosol properties from satellite measurements of backscattered ultraviolet radiation: Theoretical basis, J. Geophys. Res., 103(D14), 17,099–17,110, doi:10.1029/98JD00900.

Twomey, S.: The nuclei of natural cloud formation part I: The chemical diffusion method and its application to atmospheric nuclei, Pure Appl. Geophys., 43, 227–242, doi:10.1007/BF01993559, 1959

J.P. Veefkind (ed.) and co-authors (2008), Technical Report: TN-CAM-KNMI-050, CAMELOT Final Report.

Waquet, F., J. Riedi, L. C.-Labonnote, P. Goloub, B. Cairns, J.-L. Deuzé, and D. Tanré, 2009b: Aerosol remote sensing over clouds using A-Train observations. J. Atmos. Sci. 66, 2468–2480.

Winker, D. M., J. Pelon, J. A. Coakley Jr., S. A. Ackerman, R. J. Charlson, P. R. Colarco, P. Flamant,Q. Fu, R. M. Hoff, C. Kittaka, T. L. Kubar, H. Le Treut, M. P. McCormick, G. Mégie, L. Poole, K. Powell, C. Trepte, M. A. Vaughan, B. A. Wielicki, 2010: The CALIPSO Mission: a global 3D view of aerosols and clouds. Bull. Amer. Meteorol. Soc. 91, 1211–1229.

Wu, L., Hasekamp, O., van Diedenhoven, B., and Cairns, B.: Aerosol retrieval from multiangle, multispectral photopolarimetric measurements: importance of spectral range and angular resolution, Atmos. Meas. Tech., 8, 2625-2638, doi:10.5194/amt-8-2625-2015, 2015.

Zhu, L., J. V. Martins, and L. A. Remer (2011), Biomass burning aerosol absorption measurements with MODIS using the critical reflectance method, J. Geophys. Res., 116, D07202, doi:10.1029/2010JD015187

9 APPENDIX: REFERENCE LER SPECTRUM FOR SNR

*Table 4: Lambertian Equivalent Reflectance (LER) values for solar zenith angle (SZA) of 70°, for the Signal to Noise Ratio (SNR) requirement. The radiance at top of atmosphere is given by F_0 * COS(SZA) / PI, F_0 being the solar flux perpendicular to the solar beam.*

wavelength [nm]	**LER**
0.370000E+03	0.279702E+00
0.380000E+03	0.262193E+00
0.390000E+03	0.245808E+00
0.400000E+03	0.230453E+00
0.410000E+03	0.216086E+00
0.420000E+03	0.202657E+00
0.430000E+03	0.190108E+00
0.440000E+03	0.178397E+00
0.450000E+03	0.167469E+00
0.460000E+03	0.157277E+00
0.470000E+03	0.147768E+00
0.480000E+03	0.138892E+00
0.490000E+03	0.130603E+00
0.500000E+03	0.122865E+00
0.510000E+03	0.115631E+00
0.520000E+03	0.108864E+00
0.530000E+03	0.102528E+00
0.540000E+03	0.965891E-01
0.550000E+03	0.910137E-01
0.560000E+03	0.857729E-01
0.570000E+03	0.808415E-01
0.580000E+03	0.761948E-01
0.590000E+03	0.718103E-01
0.600000E+03	0.676608E-01
0.610000E+03	0.637415E-01
0.620000E+03	0.600267E-01
0.630000E+03	0.564968E-01
0.640000E+03	0.531448E-01
0.650000E+03	0.499532E-01
0.660000E+03	0.469097E-01
0.670000E+03	0.440029E-01
0.680000E+03	0.412225E-01
0.690000E+03	0.400000E-01
0.700000E+03	0.400000E-01
0.710000E+03	0.400000E-01
0.720000E+03	0.400000E-01
0.730000E+03	0.400000E-01

0.740000E+03	0.400000E-01
0.750000E+03	0.400000E-01
0.760000E+03	0.400000E-01
0.770000E+03	0.400000E-01